U.S. DEPARTMENT OF COMMERCE
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PB-280 482

Contaminants Associated With Direct and Indirect Reuse of Municipal Wastewater

SCS Engineers, Long Beach, Calif

Prepared for

Health Effects Research Lab, Cincinnati, Ohio

Mar 78

PB 280 482

EPA-600/1-78-019
March 1978

Environmental Health Effects Research Series

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Health Effects Research Laboratory
Office of Research and Development
U.S. Environmental Protection Agency
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TECHNICAL REPORT DATA

(Please read Instructions on the reverse before completing)

1. REPORT NO. EPA-600/1-78-019	2.	3. RECIPIENT'S ACCESSION NO. PB280482
4. TITLE AND SUBTITLE Contaminants Associated with Direct and Indirect Reuse of Municipal Wastewater		5. REPORT DATE March 1978 issuing date
		6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S)		8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS SCS Engineers, Inc. 4014 Long Beach Boulevard Long Beach, California 90807		10. PROGRAM ELEMENT NO. 1CC614
		11. CONTRACT/GRANT NO. 68-02-2257
12. SPONSORING AGENCY NAME AND ADDRESS Health Effects Research Lab-Cincinnati, OH Office of Research and Development U.S. Environmental Protection Agency Cincinnati, Ohio 45268		13. TYPE OF REPORT AND PERIOD COVERED Final, 7/1/76 - 12/1/77
		14. SPONSORING AGENCY CODE EPA/600/10

15. SUPPLEMENTARY NOTES

16. ABSTRACT

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17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Waste water Waste treatment Potable water Public health Water	Drinking water Waste water reuse Water treatment Water Supply	68D

18. DISTRIBUTION STATEMENT Release to public	19. SECURITY CLASS *(This Report)* Unclassified	21. NO. OF PAGES 359
	20. SECURITY CLASS *(This page)* Unclassified	22. PRICE A16 - A01

EPA Form 2220-1 (9-73)

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EPA-600/1-78-019
March 1978

CONTAMINANTS ASSOCIATED WITH DIRECT AND INDIRECT REUSE OF MUNICIPAL WASTEWATER

by

SCS Engineers
Long Beach, California 90807

Contract No. 68-02-2257

Project Officer

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This report has been reviewed by the Health Effects Research Laboratory, U.S. Environmental Protection Agency, and approved for publication. Approval does not signify that the contents necessarily reflect the views and policies of the U.S. Environmental Protection Agency, nor does mention of trade names or commercial products constitute endorsement or recommendation for use.

FOREWORD

The U.S. Environmental Protection Agency was created because of increasing public and government concern about the dangers of pollution to the health and welfare of the American people. Noxious air, foul water, and spoiled land are tragic testimony to the deterioration of our natural environment. The complexity of that environment and the interplay between its components require a concentrated and integrated attack on the problem.

Research and development is that necessary first step in problem solution and it involves defining the problem, measuring its impact, and searching for solutions. The primary mission of the Health Effects Research Laboratory in Cincinnati (HERL) is to provide a sound health effects data base in support of the regulatory activities of the EPA. To this end, HERL conducts a research program to identify, characterize, and quantitate harmful effects of pollutants that may result from exposure to chemical, physical or biological agents found in the environment. In addition to valuable health information generated by these activities, new research techniques and methods are being developed that contribute to a better understanding of human biochemical and physiological functions, and how these functions are altered by low-level insults.

This report presents the state of knowledge concerning levels, removals, and health effects of contaminants associated with direct and indirect reuse of municipal wastewater for potable purposes. With a better understanding of the degree of insult in our drinking water, measures may be developed to overcome some of these potentially harmful materials.

R. J. Garner
Director
Health Effects Research Laboratory

PREFACE

From time immemorial man has used, with varying consequences, water contaminated by other men, by animals, and by natural processes. In modern times, it has become increasingly necessary to reuse water as populations and productivity have multiplied and limited water supplies have been used up. However, this reuse has usually been done on an unplanned basis. It has been conservatively estimated that, at the present time, approximately one-third of the population in the United States derives water from sources which are degraded to some extent by wastewater discharges. Excepting the transmission of infectious diseases, little concern - until very recently - has been given to the disease-producing potential of such contamination. It is now necessary to review the health effects of this situation in a more comprehensive fashion.

Since the public health disaster of Minimata, Japan, in the late 1950's and early 1960's (caused by ingestion of shellfish contaminated with methyl mercury) there has been a great surge in research concerning environmentally induced health effects on man. It is now generally accepted that the myriad of contaminants which are continuously discharged to the environment may produce both acute and chronic repercussions on public health through daily ingestion of air, water, and food. Recently, some forms of cancer, once thought to be of genetic etiology, have been projected to be caused or stimulated by environmental contaminants.

One area of critical consideration to public health is the direct or indirect reuse of municipal wastewater for potable purposes. Municipal wastewater systems have been the repositories of virtually every chemical contaminant known to be produced by man. The very nature of municipal wastewater streams makes them of critical importance when considering the environmentally induced health effects on man. Many questions must be clearly answered before a complete understanding of the situation will occur: What are the harmful constituents of municipal wastewater? How well does our present wastewater treatment technology remove these constituents? What eventually happens to them in the environment? How well do our water treatment plants perform in providing contaminant removals and a last line of protection? What are the effects of the contaminants on the health and well-being of man? This report is an attempt to assemble and evaluate the existing literature pertinent to these questions. It is not an in-depth study. Rather, it is intended to serve as a basis for determining the health risks involved and the future research requirements for direct and indirect potable water reuse.

For purposes of this report, direct reuse is defined as the discharge of the treated municipal wastewater directly into a raw water supply without intervening travel, and dilution in natural surface or groundwater. No such direct reuse currently exists in the United States; however, direct reuse has been practiced in South Africa for about ten years, and long-range plans for direct reuse are being considered in some United States municipalities (e.g., Denver).

Indirect reuse is defined as the reuse of treated municipal wastewater as a raw water supply after the wastewater has entered, comingled, and essentially become a part of a natural surface or groundwater resource. A significant percentage of the nation's raw water supply is derived from surface waters such as major lakes and rivers, and consists in part of treated wastewater from other municipalities. This indirect reuse has long been accepted by the public and the waterworks industry as normal and inevitable. Indirect reuse also includes introduction of treated wastewater into groundwater aquifers through percolation or well injection. This practice is often labeled groundwater recharge and may be a formal, intentional program, or simply a result of land disposal of wastewater.

Obviously, in some cases there is only a fine line separating direct reuse from indirect reuse. For example, if a municipal water department owns and operates a large raw water storage reservoir and treated wastewater is introduced into that reservoir, even as a relatively small percentage of the "fresh" water volume, that is considered direct reuse. Conversely, a large volume of the wastewater in a river in the Midwest may come from wastewater from upstream municipalities, and yet a water supply taken from that river would be considered indirect reuse. Advocates of direct reuse point to comparisons of this type to show that the stigma attached to well-designed direct reuse is irrational. Opponents of direct reuse answer that "two wrongs don't make a right," and that insufficient knowledge exists about both direct and indirect reuse to ensure a "safe" water supply for the public.

ABSTRACT

This report is an attempt to compile the published quantitative data available concerning the health effects associated with direct and indirect reuse of treated municipal wastewater for potable purposes. The assembled information includes data on the effectiveness of conventional water and wastewater treatment and disposal operations in reducing public health contaminant concentrations, as well as data on the transport of these contaminants through the environment back to man. The data have been organized in such a manner that the various pathways of pollutants to man can be evaluated for relative public health significance in order to establish necessary research priorities.

Wastewater treatment processes evaluated include conventional secondary treatment and tertiary processes. Wastewater disposal techniques evaluated include direct discharge to fresh surface waters and land application. Water treatment processes evaluated include conventional treatment (chemical coagulation, with or without filtration, and disinfection) and advanced water treatment (carbon adsorption, ion exchange, and reverse osmosis). A discussion of public health considerations is also included.

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SECTION 1

INTRODUCTION

OBJECTIVES

The purpose of this report is to clearly describe the state-of-the-art knowledge pertinent to potential health effects from direct and indirect reuse of treated municipal wastewater for potable purposes. No new basic research is intended. Rather, the report ties together the results of past and ongoing research:

- The ranges of concentrations of contaminants contained in the influent to municipal wastewater treatment plants;
- The effectiveness of conventional wastewater treatment processes in the removal, modification, or inactivation of these contaminants;
- The potential hazards associated with effluent disposal practices in regard to the introduction of contaminants to the ecosystem;
- Pathways by which contaminants associated with wastewater treatment/disposal operations can intersect with man;
- The effectiveness of conventional water treatment processes in the removal, modification, or inactivation of these contaminants;
- The chronic and acute effects of the contaminants within the human body.

SCOPE

This report compiles the published quantitative data available concerning the potential health effects associated with direct and indirect reuse of treated municipal wastewater for potable purposes. The assembled information includes data on the effectiveness of conventional water and wastewater treatment and disposal operations in reducing public health contaminant concentrations, as well as data on the transport of these contaminants through the environment back to man. The data have been organized in such a manner that the various pathways of pollutants to man can be evaluated for relative public health significance in order to establish necessary research priorities.

Wastewater treatment processes evaluated include conventional secondary treatment and tertiary processes. Wastewater disposal techniques evaluated include direct discharge to fresh surface waters and land application. Water treatment processes evaluated include conventional treatment (chemical coagulation, with or without filtration, and disinfection) and advanced water treatment (carbon adsorption, ion exchange, and reverse osmosis).

APPROACH

This project was accomplished in three distinct phases or tasks:

Task I - Literature Review

The first task reviewed the present literature, providing information and data relevant to the health effects associated with the direct and indirect reuse of municipal wastewater for potable purposes. The relevant literature included the transport and losses of the various contaminants through the various pathways and unit operations between the raw waste and usable drinking water.

Library Research--

Research assistants obtained copies of pertinent literature from several major university libraries, the Library of Congress, information retrieval systems (e.g., NTIS), and the SCS Engineers in-house library. Due to the breadth of the topic, several constraints were placed upon the literature search in order to assure a workable, yet comprehensive, volume of information. Major constraints follow:

- Only literature published within the last ten years was reviewed, except in unusual circumstances.

- Except in unusual circumstances, only literature directly addressing wastewater and water treatment and disposal, and health effects was reviewed. There is, of course, a large volume of related literature (e.g., biological, medical, meteorological) that could provide additional insights; however, this peripheral literature was excluded from the investigation.

Correspondence--

To obtain the very latest scientific information available, as well as to find areas in which research is presently being conducted, letters were sent soliciting literature and data from the following information sources:

- State regulatory and public health agencies;
- Major sanitary districts known to be conducting research; and
- Universities conducting related research.

The responses from these sources have been incorporated into the project.

Task II - Data Analysis

The data and information obtained in Task I were organized and analyzed to trace the movements and losses of the various contaminants through the pathways associated with direct and indirect reuse of municipal wastewater. These pathways included the waste treatment system, any applicable environmental pathways such as river transport or groundwater, and the potable water treatment plant. The potential health significance of this transport of the contaminants was delineated where information was available.

Task III - Report Preparation

This report is organized by pathway, rather than by contaminant, so that various public health impairing contaminants can be traced through wastewater treatment plants, through the biosphere, and finally back to man. Figure 1 illustrates the alternate pathways that various contaminants may follow to reach man after leaving the wastewater treatment plant. In dark relief are those pathways associated with direct and indirect reuse for potable purposes.

Each major wastewater and water treatment step and disposal method is treated in a separate subsection of the report. The report does not, however, contain a separate section for each contaminant. If the reader is interested in mercury, for example, he must skip through the report and read about the fate of mercury during individual treatment processes, when released to the biosphere, and the consequent health effects on man. To facilitate the task of tracing a single contaminant through the report, each section is divided into subsections by contaminant type.

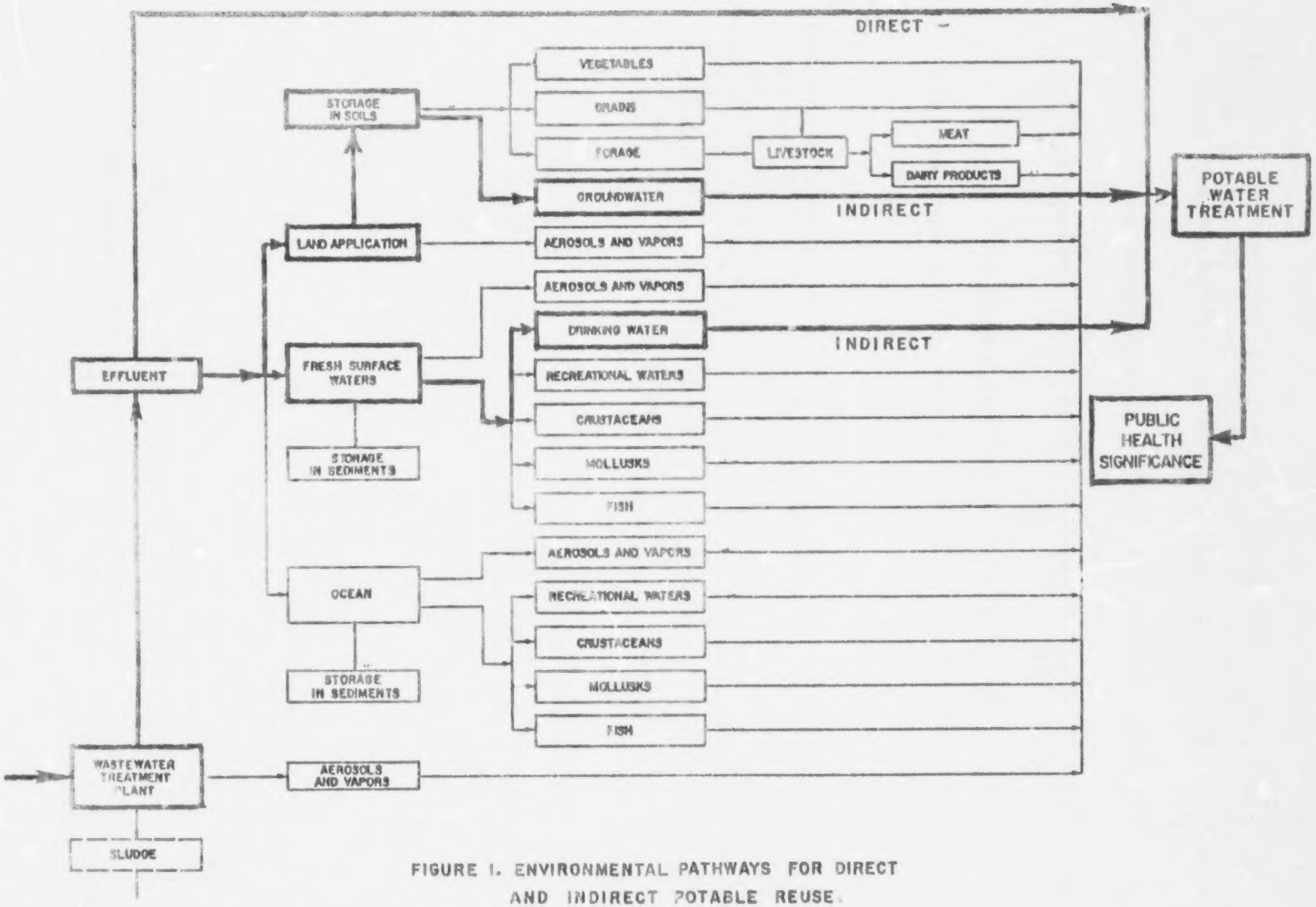

DIRECT
WASTEWATER TREATMENT PLANT
SLUDGE
EFFLUENT
AEROSOLS AND VAPORS
STORAGE IN SOILS
LAND APPLICATION
FRESH SURFACE WATERS
STORAGE IN SEDIMENTS
OCEAN
STORAGE IN SEDIMENTS
VEGETABLES
GRAINS
FORAGE
GROUNDWATER
AEROSOLS AND VAPORS
AEROSOLS AND VAPORS
DRINKING WATER
RECREATIONAL WATERS
CRUSTACEANS
MOLLUSKS
FISH
AEROSOLS AND VAPORS
RECREATIONAL WATERS
CRUSTACEANS
MOLLUSKS
FISH
LIVESTOCK
MEAT
DAIRY PRODUCTS
INDIRECT
INDIRECT
POTABLE WATER TREATMENT
PUBLIC HEALTH SIGNIFICANCE

FIGURE 1. ENVIRONMENTAL PATHWAYS FOR DIRECT AND INDIRECT POTABLE REUSE.

The selection of wastewater contaminants for consideration in this study was difficult, because of the many public health impairing constituents found in wastewater. Several of the traditional wastewater parameters (BOD, suspended solids, etc.) pose no direct threat to public health, although some of the direct health-impairing contaminants may be associated with these traditional wastewater parameters. Therefore, some of the traditional parameters are included.

Within each section of this report (which traces wastewater contaminants through municipal wastewater treatments, through the environment, and through water treatment plants, and which details the epidemiological and pathological effects on man) information is organized and presented in the following contaminant groups:

1. Water Quality Parameters
2. Elemental Contaminants
3. Biocidal Contaminants
4. Synthetic/Organic Contaminants
5. Biological Contaminants

The water quality parameters group contains those water measurements that are traditionally associated with wastewater treatment systems, as well as those contaminants that do not readily fit in any other category. Table 1 lists these water quality parameters. Most of the parameters listed in this table do not pose a direct threat to public health, but may be related to public health impairing contaminants. For example, heavy metals or viruses may be adsorbed to the surface of suspended solids and may be transported through the wastewater treatment plant and biosphere in this manner. However, the nitrogenous compounds (ammonia, nitrates, and nitrites) may directly threaten public health.

TABLE 1 . WATER QUALITY PARAMETERS

1. Ammonia
2. Biochemical Oxygen Demand (BOD)
3. Chemical Oxygen Demand (COD)
4. Nitrates
5. Nitrites
6. Phosphates
7. Suspended Solids
8. Total Dissolved Solids (TDS)
9. Total Organic Carbon (TOC)

The elemental contaminants group, consists of the ions, compounds, and complexes of the metals and metalloids listed in Table 2. Many of these contaminants are required in trace quantities for normal human metabolic functions, yet higher levels of these trace elements may cause significant health problems.

TABLE 2 . ELEMENTAL CONTAMINANTS

1.	Aluminum	9.	Cobalt	17.	Nickel
2.	Antimony	10.	Copper	18.	Selenium
3.	Arsenic	11.	Germanium	19.	Thorium
4.	Barium	12.	Iron	20.	Tin
5.	Beryllium	13.	Lead	21.	Uranium
6.	Boron	14.	Manganese	22.	Zinc
7.	Cadmium	15.	Mercury		
8.	Chromium	16.	Molybdenum		

Biocidal contaminants are those contaminants normally used to control insect or disease vectors. Table 3 lists these biocidal contaminants. DDT, DDD, DDE, aldrin, dieldrin, and endrin are all chlorinated hydrocarbons; they were considered separate from the chlorinated hydrocarbon classification because the literature often dealt with these specific pesticides as individual entities.

TABLE 3 . BIOCIDAL CONTAMINANTS

1.	DDT	7.	Chlorinated hydrocarbons
2.	DDD	8.	Arsenated hydrocarbons
3.	DDE	9.	Organonitrogen pesticides
4.	Aldrin	10.	Organophosphorus pesticides
5.	Dieldrin	11.	Herbicides
6.	Endrin	12.	Soil sterilants

The synthetic/organic contaminants group includes many synthetically produced organic chemicals that have found their way into water systems. For the purposes of this report, information was gathered on any synthetic/organic chemicals in wastewater treatment processes or the biosphere. A selection of such contaminants, recently identified by the Environmental Protection Agency in an evaluation of organic compounds in the New Orleans area water supply are listed in Table 4.

Traditionally, biological contaminants have received the most attention in wastewater treatment, since these contaminants may directly cause infection in the consumer. Biological contaminants considered in this study are listed in Table 5.

Table 4. ORGANIC COMPOUND IDENTIFICATION
NEW ORLEANS AREA WATER SUPPLY STUDY (615)

1. Acetaldehyde
2. Acetone
3. Alkylbenzene-C_2 isomer
4. Alkylbenzene-C_2 isomer
5. Alkylbenzene-C_2 isomer
6. Alkylbenzene-C_3 isomer
7. Alkylbenzene-C_3 isomer
8. Alkylbenzene-C_3 isomer
9. Atrazine (2-chloro-4-ethylamino-6-isopropylamino-*s*-triazine)
10. Deethylatrazine (2-chloro-4-amino-6-isopropylamino-*s*-triazine)
11. Benzyl butyl phthalate
12. Bromodichloroethane
13. Bromoform
14. Butanone
15. Carbon disulfide
16. Carbon tetrachloride
17. bis-2-Chloroethyl ether
18. Chloroform
19. bis-2-Chloroisopropyl ether
20. n-Decane
21. Decane-branched isomer
22. Dibromodichloroethane isomer
23. Dibromochloromethane
24. Dibutyl phthalate
25. 2,6-Di-t-butyl-*p*-benzoquinone
26. Dichlorobenzene isomer
27. 1,2-Dichloroethane
28. Dichloromethane
29. Dieldrin
30. Diethyl phthalate
31. Di(2-ethylhexyl) phthalate
32. Dihexyl phthalate
33. Dihydrocarvone
34. Diisobutyl phthalate
35. Dimethyl phthalate
36. Dioctyl adipate
37. Dipropyl phthalate
38. n-Dodecane
39. Endrin
40. Ethanol
41. *o*-Ethyltoluene
42. *p*-Ethyltoluene

TABLE 4 (continued)

43. 1, 2, 3, 4, 5, 7, 7-Heptachloronorbornene

44. Heptachloronorbornene isomer

45. Hexachloro-1,3-butadiene

46. Hexachloroethane

47. Isophorone

48. Limonene

49. Methanol

50. Methylbenzoate

51. 3-Methylbutanal

52. 2-Methylpropanal

53. n-Nonane

54. n-Pentadecane

55. Tetrachloroethane isomer

56. Tetrachloroethylene

57. n-Tetradecane

58. Toluene

59. 1,1,2-Trichloroethane

60. 1,1,2-Trichloroethylene

61. n-Tridecane

62. Trimethyl-trioxo-hexahydrotriazine isomer

63. Triphenyl phosphate

64. n-Undecane

65. Undecane-branched isomer

TABLE 5. BIOLOGICAL CONTAMINANTS

1. Adenovirus
2. *Clostridium botulinum*
3. *Clostridium perfringens*
4. Coliforms
5. Coxsackie virus (A&B)
6. ECHO virus
7. *Erysipelothiix rhusiopathie*
8. *Escherichia coli*
9. Fecal streptococci
10. *Francisella tularensis*
11. Hepatitis virus
12. Leptospira
13. *Listeria monocytogenes*
14. Mycobacterium
15. Parasitic worms
16. Polio virus
17. Protozoa
18. Salmonella
19. Shigella
20. *Staphylococcus aureus*
21. Vibrio cholerae
22. Yeasts

SECTION 2

WASTEWATER INPUTS

INTRODUCTION

Untreated wastewater input composition is the first point of interest in determining the pathways pollutants may follow from wastewater management systems back to man in direct and indirect reuse situations. In addition to domestic sewage, input sources may include various industrial wastes, storm water, and groundwater infiltration, in various combinations. Moreover, the input proportions of an individual system change with time; pollutant concentrations and volumes vary hourly, daily, weekly, and seasonally.

Research surveyed regarding input concentrations to municipal treatment facilities is presented in Table 6 . Substantial literature concerning input compositions is available for water quality parameters and elemental and biological contaminants. With few exceptions, however, input concentrations of biocidal and synthetic-organic contaminants have not been investigated. It was therefore difficult to determine the effect of subsequent treatment processes upon such contaminants. Regulations for industrial discharges of complex mixtures of organic compounds generally only require reporting of the BOD or COD, suspended solids, and similar water quality parameters. In many instances, industrial concerns themselves may not know the detailed composition of their waste streams.

Since 1972, the development of the National Pollution Discharge Elimination System (NPDES) permit programming has changed the typical input composition to municipal sanitary systems through increased restrictions on inputs from industrial waste. In particular, these restrictions have reduced the amount of heavy metal contaminants entering municipal treatment systems.

Municipal treatment systems that process only domestic wastes and that have no excessive infiltration show predictable diurnal and seasonal patterns. However, most urban sanitary systems and many rural systems also contain industrial wastes. Composition and flow volumes may vary significantly as a function of these industrial inputs. Food processing plants can

TABLE 6. LITERATURE REVIEWED PERTAINING TO THE COMPOSITION OF WASTEWATER INPUTS TO MUNICIPAL TREATMENT SYSTEMS

Contaminant	Reference Number
Water Quality Parameters	
Ammonia	22, 90, 273, 368, 390, 406, 450, 503, 516, 582, 647, 651, 700
BOD	19, 69, 171, 251, 273, 302, 345, 368, 390, 450, 503, 526, 569, 647, 651, 653, 700
COD	19, 69, 134, 171, 251, 312, 390, 391, 450, 516, 526, 582, 647, 651, 700
Chlorides	19, 22, 161, 450, 516
Cyanides	390
Fluorides	390
Nitrates	22, 90, 273, 368, 406, 450, 503, 510, 516, 582, 647, 651
Nitrites	90, 273, 368, 390, 406, 450, 503, 516, 582, 647, 651, 700
Oil & grease	541
Phosphates	19, 22, 69, 132, 134, 161, 273, 304, 368, 390, 406, 450, 516, 582, 647, 651, 700
Suspended solids	22, 134, 171, 312, 368, 516, 526, 582, 647, 651, 653, 700
Total dissolved solids	1, 390, 647
Total organic carbon	69, 134, 312, 516, 647
Other (general)	7, 312
Elemental Contaminants	
Aluminum	26, 134, 390, 538
Arsenic	19, 390

TABLE 6 (continued)

Contaminant	Reference Number
Barium	19, 390
Cadmium	19, 37, 159, 390, 391, 471, 702
Chromium	19, 134, 159, 390, 391, 462, 471, 473, 702
Cobalt	134, 273, 390, 471, 702
Copper	19, 107, 134, 159, 273, 390, 471, 702
Iron	26, 134, 273, 450, 471, 538, 702
Lead	19, 107, 134, 390, 471, 493
Manganese	134, 273, 390, 450, 471, 702
Mercury	19, 392, 471
Molybdenum	390, 702
Nickel	107, 159, 273, 390, 471, 702
Selenium	19, 390, 391
Tin	390
Zinc	19, 26, 107, 134, 159, 390, 702
Synthetic/Organic Contaminants	317, 434, 702
Biological Contaminants	
Coliforms	134, 163, 251, 368, 390, 526, 560
ECHO virus	560
Fecal streptococci	163, 236, 390, 560
Mycobacterium	223
Parasitic worms	223
Polio virus	560
Protozoa	223

TABLE 6 (continued)

Contaminant	Reference Number
Salmonella	223, 339
Shigella	223
Virus	198, 223

contribute seasonally high BOD discharges; large metal platers and metal finishers may periodically contribute high metal waste concentrations. Certain situations require particular attention when evaluating potential adverse effects on human health, including:

- Large hospital complexes connected to small sanitary systems;
- Situations in which large amounts of waste from metal platers and metal finishers discharge to the system;
- Tanneries discharging to the system;
- Petrochemical and related complexes;
- Biocide manufacturing facilities;
- Specialty chemical manufacturing or formulating facilities; and
- Combined or heavily infiltrated systems that bypass excess flow without adequate treatment.

WATER QUALITY PARAMETERS

The water quality parameters that characterize wastewater entering into a sanitary treatment plant have been extensively studied. Concentration ranges recorded in the reviewed literature are presented in Table 7 . Atypical situations that lie outside these ranges are known to occur under special conditions. Such exceptions can usually be attributed to excessive industrial inputs, excessive infiltration, or unusual characteristics of the fresh-water supply to the service area.

Diurnal flow patterns reported by Nomura and Young (486) show that suspended solids concentrations in raw sewage are directly related to sewage flow rates. The suspended solids content of the raw sewage studied ranged from 20 mg/ℓ in the late night flow (attributed to infiltration) up to a high of 350 mg/ℓ during the peak daily sewage flow rate.

Storm and municipal systems show considerable variation in flow rate as a function of time; consequently, contaminant concentrations vary greatly (7, 368, 502). Water quality parameter variations due to storm system input will be a function of:

- Normal concentrations during dry weather flow
- Amount and duration of precipitation
- Time since the last rain
- Season

TABLE 7. SUMMARY OF WATER QUALITY PARAMETERS CHARACTERIZING WASTEWATER INPUT TO MUNICIPAL TREATMENT FACILITIES*

Constituent	Range, mg/ℓ
Ammonia nitrogen	8 to 50
Total nitrogen	20 to 85
Organic-nitrogen	5 to 32
Nitrate-nitrogen	0 to 3
Nitrite-nitrogen	0 to 1
Chloride	25 to 203
Oil and grease	1 to 50
Total phosphorus	4 to 50
Phosphate as PO_4	5 to 50
Inorganic phosphorus	8 to 13
Organic phosphorus	1 to 5
BOD	30 to 600
COD	100 to 1000
Suspended solids	30 to 350
Total dissolved solids	250 to 1400

*As reported in the literature reviewed.

ELEMENTAL CONTAMINANTS

The type and amount of elemental contaminants contained in any treatment system input will depend primarily upon the type and amount of industrial wastes entering that system. A great deal of material is available on industrial discharges to municipal systems. A summary range of elemental influent characteristics as reported in the literature is presented in Table 8. The Interim Drinking Water Standards have been included to provide a standard of comparison.

The literature reviewed did not provide a comprehensive survey of the sources of these metallic contaminants, but the following factors appear to be important:

- Input water composition
- Input water scaling and corrosion potential
- Type and age of domestic water piping systems
- Type and amount of industrial discharges
- Type of municipal system (combined or separate storm water).

The relative significance of these factors will again depend upon the specific site. The results of a study by Davis and Jacknow (159) of the 12 municipal treatment plants in New York City (presented in Table 9) give some idea of the ratio of contributions from various sources that might be expected in a large urban area. This same study contains source breakdowns for individual plants. Input from the residential sector contained concentrations exceeding the Interim Primary Drinking Water Standards by a factor of 2.

A knowledge of the physical and chemical forms of elemental contaminants is necessary in order to adequately evaluate potential public health effects. In particular, it is necessary to know whether these elements are present as particulate materials or soluble species. Data of this type are apparently unavailable at this time.

BIOLOGICAL CONTAMINANTS

Information available in the literature concerning input concentrations of biological contaminants generally addresses primary indicator organisms rather than specific pathogens. A summary range of biological contaminants reported in this literature is presented in Table 10.

TABLE 8. SUMMARY OF THE CONCENTRATION RANGES OF ELEMENTAL CONTAMINANTS FOUND IN WASTEWATER INPUTS TO MUNICIPAL TREATMENT SYSTEMS*

Constituent	Range (mg/ℓ)	Interim Drinking Water Standards
Aluminum	0.3 to 3.0	--
Antimony	No data in literature	--
Arsenic	0 to 0.02	.05
Barium	0 to 0.02	1
Beryllium	No data in literature	--
Boron	0.5 to 3	--
Cadmium	0.01 to 0.2	.01
Chromium	0.01 to 0.3	.05
Cobalt	No data in literature	--
Copper	0.01 to 0.5	--
Germanium	No data in literature	--
Iron	0.5 to 6.5	--
Lead	0 to 1	.05
Manganese	0.05 to 0.15	--
Mercury	0.0002 to 0.003	.002
Molybdenum	No data in literature	--
Nickel	0.05 to 0.5	--
Selenium	0 to 0.11	.01
Thorium	No data in literature	--
Tin	No data in literature	--
Uranium	No data in literature	--
Zinc	0.01 to 2.10	--

*As reported in the literature reviewed.

TABLE 9. AVERAGE HEAVY METAL LOADINGS AND PROBABLE SOURCES FOR TWELVE NEW YORK CITY MUNICIPAL TREATMENT PLANTS (159)

Metals	Total Loading (lb/d)	Electroplating and Photoengraving (lb/d)	Percentage of total	Estimate of Residential Contribution (lb/d)	Percentage of Total	Other Industries, Urban Storm water, and Other Contributions (lb/d)	Percentage of total
Copper	3,820	611	15	1,440	38	1,770	46
Chromium	2,340	345	14	640	27	1,350	58
Nickel	1,870	1,010	53	640	34	220	12
Zinc	10,440	830	7	1,680	16	7,930	76
Cadmium	341	65	19	128	38	148	43

TABLE 10 . SUMMARY OF THE CONCENTRATION RANGES OF BIOLOGICAL CONTAMINANTS FOUND IN WASTEWATER INPUTS TO MUNICIPAL TREATMENT SYSTEMS*

Contaminant	Range/100 mℓ
Total coliforms	1×10^6 to 4.6×10^7
Fecal coliforms	3.4×10^5 to 4.9×10^7
Fecal streptococci	6.4×10^4 to 4.5×10^6
Virus	5 to 100,000 virus units/ℓ

*As reported in the literature reviewed.

Pound and Crites (526) reported that raw municipal sewage contains from 10^6 to 10^8 total coliforms and from 480 to 1,677 plaque-forming units (pfu)/ℓ of enteric viruses. The average enteric virus density in domestic sewage reported by the ASCE (198) was approximately 500 virus units/100 mℓ. Coliform densities averaged 4.6×10^7/100 mℓ. Analyses of wastewater reported by Kampelmacher and Jansen (339) show that salmonella is regularly present.

Viral concentrations in raw wastewater reported by Foster and Engelbrecht (223) range from 5 to 100,000 viral pfu/ℓ, with reported seasonal variations of a mean of 4,000 pfu/ℓ during warm months and a mean of 200 pfu/ℓ during cold months. However, no universal procedure or system is presently available for cultivation of all viruses. It is likely, therefore, that many of the investigations of virus density in wastewater have not included all viruses present due to the selectivity of techniques employed.

Fecal streptococci contaminants present in the domestic wastewater of seven communities were studied by Geldreich and Kenner (236). Figures obtained from this study are presented in Table 11.

TABLE 11. DISTRIBUTION OF FECAL STREPTOCOCCI IN DOMESTIC WASTEWATER AND STORMWATER RUNOFFS (236)

Water Source	Densities/100 ml				Occurrence (%)			
	Fecal Coliforms	Fecal Streptococci	Ratio FC/FS	Total Strains Examined	Entero-cocci	*S.bovis* *S.equinus*	Atypical *S.faecalis*	*S.faecalis liquifaciens*
Domestic wastewater								
Preston, Ida.	340,000	64,000	5.3	39	79.5	None	None	20.5
Fargo, N. D.	1,300,000	290,000	4.5	50	100.0	None	None	None
Moorehead, Minn.	1,600,000	330,000	4.9	50	90.0	10.0	None	None
Cincinnati, Ohio	10,900,000	2,470,000	4.4	428	71.5	2.8	1.6	24.1
Lawrence, Mass.	17,900,000	4,500,000	4.0	50	84.0	4.0	None	12.0
Monroe, Mich.	19,200,000	700,000	27.9	70	78.6	1.4	4.3	15.7
Denver, Colo.	49,000,000	2,900,000	16.9	70	85.7	11.4	2.9	None
Storm water								
Business district	13,000	51,000	0.26	1,476	78.5	1.6	1.2	18.8
Residential	6,500	150,000	0.04	1,158	80.0	0.5	1.4	18.1
Rural	2,700	58,000	0.05	445	87.4	0.5	0.2	11.9

SECTION 3

PRIMARY TREATMENT

INTRODUCTION

Primary treatment is intended to physically remove settleable solids and most of the discrete suspended and floating solids from the municipal wastewater stream preparatory to secondary treatment. In addition, primary treatment removes a limited portion of the soluble constituents.

In primary treatment, the wastewater influent is divided into three output pathways: primary effluent, primary sludge (including grit, screenings, and precipitated matter), and aerosols. Effluent from primary treatment can be directly discharged (until 1977), discharged after disinfection (again until 1977), or treated by a secondary process. Primary sludge is normally subject to additional processing. At some ocean coastal sites, however, the sludge is discharged without further treatment. Aerosols are rarely a problem, due to the absence of excessive turbulence.

The information reviewed during this study is tabulated in Table 12. Most research conducted to date concerns the removal by primary treatment of water quality parameter constituents. It is only in recent years that researchers have examined the effect of primary treatment on various public health impairing contaminants.

WATER QUALITY PARAMETERS

For several decades, extensive work has been reported concerning general contaminant removal efficiencies for primary treatment. Removal varies widely, depending upon the physical and chemical characteristics of the wastewater, the proportion of settleable solids, concentrations of the solids, and detention time. Mitchell (453) reported general performance efficiencies that could be expected for typical primary treatment. Removals achieved during a three-year study of an operational primary treatment system are shown in Table 13.

TABLE 12. LITERATURE REVIEWED PERTAINING TO PRIMARY WASTEWATER TREATMENT

Contaminant	Reference Number
Water Quality Parameters	
Ammonia	453
BOD	206, 318, 453, 654, 665, 707
COD	251, 318, 443, 453, 665
Oil and grease	251, 453
Phosphates	443
Suspended solids	206, 251, 318, 368, 443, 453, 665, 707
Total organic carbon	251
Other (general)	280, 318
Elemental Contaminants	
Arsenic	453
Cadmium	124, 453, 507
Copper	124, 443, 453, 507
Iron	124, 443
Lead	124, 453, 507
Mercury	124, 453
Nickel	124, 453, 507
Selenium	529
Zinc	124, 443, 453, 507
Biological Contaminants	
Coliforms	88, 251, 293, 357, 443, 583, 594

TABLE 12 (continued)

Contaminant	Reference
Coxsackie virus (A & B)	88
Escherichia coli	293, 443
Fecal streptococci	293
Hepatitis virus	586
Mycobacterium	88, 223
Parasitic worms	88, 217, 223
Polio virus	51, 88
Protozoa	88, 223
Salmonella	88, 217, 223, 293, 586
Shigella	88, 586
Vibrio cholerae	88
Virus	38, 48, 49, 50, 51, 54, 55, 88, 200, 223, 242, 487, 583
Other (general)	88, 586

TABLE 13. PRIMARY SETTLING TANK PERFORMANCE (453)

Parameter	Primary Influent (mg/ℓ)	Primary Effluent (mg/ℓ)	Percent Removal
COD	539	315	42
BOD_5	269	165	39
Suspended Solids	279	103	64
Oil and Grease	72	28	61
Ammonia Nitrogen	34	20	41

When used as a coagulant during primary treatment, lime is most effective in reducing certain water quality parameter constituents. When lime addition of 350 mg/ℓ was followed by air flotation, Mennell et al. (443) reported the following removals: turbidity, 98.5 percent; suspended solids, 95 percent; COD, 60 percent; and total phosphorus, 99 percent. Total nitrogen removal varied between 10 and 20 percent. Lower lime dosages provided proportionally lower removal percentages. The practice of dosing primary clarifiers with chemical coagulants will probably increase, as municipalities attempt to cost effectively meet federal and state water quality standards.

ELEMENTAL CONTAMINANTS

Recent interest in elemental contaminants, particularly trace metals, has prompted investigation into the partitioning of these contaminants in the wastewater treatment stream. Primary treatment receives elemental contaminants in a variety of forms, e.g., soluble, insoluble, and complexed. The concentrations of each form vary intrinsically as a complex function of such factors as influent metal concentration, pH, and ligand concentration.

Mitchell (453) reported removal efficiencies for primary settling over a three-year period at the Hyperion treatment plant, Los Angeles, California, as shown in Table 14.

TABLE 14. PRIMARY TREATMENT REMOVAL OF METAL ELEMENTS (453)

	Primary Influent(mg/ℓ)	Primary Effluent(mg/ℓ)	Percent Removal
Copper	0.39	0.25	36
Zinc	0.66	0.42	36
Nickel	0.30	0.24	20
Lead	0.30	0.7	--
Arsenic	0.015	0.017	--
Cadmium	0.01	0.02	--
Chromium	0.55	0.37	32

No explanation was offered to account for the anomaly of increased lead, arsenic, and cadmium concentrations.

Chen and Lockwood (124), also working with the Hyperion treatment plant, discussed the partitioning of trace metals with particulates as a function of particle size. It was reported that in primary effluent, more than 70 percent of the total trace metals content is associated with particulates as opposed to the soluble ionic form. However, only 10 to 20 percent of the Ni, Pb, and Mn was associated with the particulate fraction. Similar concentrations of metals were found on the larger (44 μm) and the smaller (0.2 μm) particles. However, particles as large as 44 μm will settle about 200 times as rapidly as 3-μm particles. More efficient removals of elemental contaminants could be expected by the use of either increased wastewater detention time, or chemical precipitants (e.g., lime) for particle coagulation. Trace metal removals reported (443) at a lime dose of 388 mg/ℓ approached 100 percent for chromium and copper, 97 percent for iron, and 94 percent for zinc. Molybdenum was not effectively removed by this treatment.

BIOLOGICAL CONTAMINANTS

Primary sedimentation usually removes less than 50 percent of coliform and pathogenic bacteria from sewage and is relatively ineffective in removing viruses and protozoa. Literature concerning the removal of water-borne pathogens by primary treatment processes reports a varying degree of efficiency, depending in part on the type of pathogen studied. Table 15 highlights the results of Bryan's investigation of pathogen survival during primary treatment (88).

In their literature review, Foster and Englebrecht (223) reported isolation of salmonella from six of seven different primary effluent samples. The raw sludge also contained members of this genus, with 19 of 20 samples tested as positive for salmonella organisms. Tubercle bacilli were reduced about 50 percent in the wastewater stream during sedimentation. It was concluded that bacterial pathogens are ineffectively removed from wastewater by primary settling; furthermore, the process produced a sludge that, without further treatment, constitutes a health hazard.

Amoebic cysts and parasitic worm ova are also ineffectively removed by primary treatment, due to their low specific densities and resulting buoyancy. Ascaris ova are an exception: Foster and Engelbrecht (223) reported 100 percent settlement of these ova into the sludge within 15 min.

Viruses, because of their size (.02 to .3 μm), are rarely removed by sedimentation, except for those that are associated with wastewater solids. The nature of the surface chemistry of

TABLE 15. SURVIVAL OF PATHOGENS DURING PRIMARY TREATMENT (88, 223)

Pathogen	% Removal
Salmonella typhi	>50
Salmonella spp.	0-15
Streptococcus faecalis	<50
Mycobacterium	48-57
Enteroviruses	no reduction
Polio viruses	no reduction
Coxsackie viruses	<50
Amoebic cysts	no effective removal
Parasitic worm ova	50-98 (223) no effective removal (88)
Ascaris ova	100

viral units suggests that adsorption depends strongly on the chemical environment, would be expected to vary according to changes in input water chemistry, and would possibly be affected by chemical additions.

The removal of viruses by primary settling has been both researched and reviewed extensively by Berg (47, 49, 51, 54). Berg described a project (49) in which only one to two-thirds of the viral particles had settled out in one day, although 75 percent of the suspended solids had settled. In this review, Berg discussed several additional studies on viral removal by primary treatment; all failed to mention detention time, and more importantly, the levels of virus in the incoming sewage were not related to those in the effluent.

Although primary settling alone will not effectively reduce the pathogen content of wastewater, dosing primary settling tanks with chemical coagulants does show some promise in this regard. Chemical precipitation, when used during primary treatment, is capable of removing as much as 99.99 percent of the virus suspended in water, effected through the formation of a coagulant-cation-virus complex. Elevated pH levels attained during lime treatment also result in substantial reductions in viral numbers (242). Lime coagulation during primary treatment brings remarkable reductions of coliform density as well. A 99.9 percent coliform reduction was measured at a lime dose of 450 mg/ℓ (443).

SECTION 4

SECONDARY TREATMENT: ACTIVATED SLUDGE

INTRODUCTION

The activated sludge process entails the growth of microorganisms in a reactor. This effects partial biological degradation of organic compounds in wastewater to simpler organic compounds, carbon dioxide, water, microorganisms, and energy (206). The basic process requires two equipment components: aeration tanks and clarifiers. Active biological sludge is separated from the effluent in a clarifier and recycled to an aeration tank.

Activated sludge, the most popular wastewater secondary treatment process, has been extensively studied, as indicated by Table 16. Most research has focused on water quality parameters such as BOD, COD, and suspended solids. The data are usually presented as percent removal, with removal efficiency determined by difference in influent and effluent concentrations. Removal of a specific contaminant can be accomplished by separation into the sludge or by degradation through biological activity. Aerosol generation from the aeration tank is also a possible contaminant pathway.

In view of possible health effects, the difference between separation and degradation can be significant. If the treatment process merely partitions a particular contaminant into the sludge or air, it remains available for migration back to man. In contrast, biological degradation can terminate the contaminant pathway or transform the potentially harmful substance into a nontoxic form. The separation and degradation components of the removal process are often not distinguished in the activated sludge literature.

WATER QUALITY PARAMETERS

Past research on activated sludge processes has concentrated on water quality parameters, with primary emphasis on BOD, COD, and suspended solids. Because of the tremendous volume of literature associated with BOD and suspended solids, and the absence of direct health effects from these pollutants, this report placed greater emphasis on literature dealing with

TABLE 16. LITERATURE REVIEWED PERTAINING TO ACTIVATED SLUDGE

Contaminant	Reference Number
Water Quality Parameters	
Ammonia	3, 7, 24, 27, 61, 103, 150, 184, 185, 190, 203, 233, 310, 314, 389, 453, 530, 620, 630
BOD	2, 7, 61, 77, 103, 108, 147, 185, 190, 289, 310, 320, 368, 389, 398, 425, 430, 453, 485, 516, 574, 622, 635, 665, 690
COD	103, 155, 190, 289, 301, 320, 338, 389, 453, 516, 543, 620, 690
Chlorides	62, 403, 446, 506
Cyanides	320, 405, 453, 506
Fluorides	506
Nitrates	7, 24, 27, 61, 67, 185, 190, 195, 231, 233, 630
Nitrites	7, 67, 185, 233, 630, 702
Oil and grease	393, 453, 506,702
Phosphates	7, 10, 26, 27, 61, 62, 103, 150, 185, 190, 195, 215, 233, 269, 301, 310, 376, 398, 446, 453, 567, 622, 625, 628
Suspended solids	2, 7, 61, 77, 103, 123, 185, 289, 320, 336, 368, 453, 516, 665, 702
Total dissolved solids	103, 147, 185, 263, 446
Total organic carbon	7, 61, 62, 185, 311, 543, 620, 702
Other (general)	203, 205, 206, 280, 302, 318, 606, 620
Elemental Contaminants	
Aluminum	269, 398, 486

TABLE 16 (continued)

Contaminant	Reference Number
Elemental Contaminants	
Arsenic	506
Barium	506
Boron	185, 446, 506
Cadmium	123, 124, 125, 453, 471, 476, 486, 506, 507, 615
Chromium	31, 33, 123, 124, 320, 453, 456, 462, 471, 506, 615
Cobalt	186, 471
Copper	31, 33, 123, 124, 125, 320, 430, 453, 471, 486, 506, 507, 615
Iron	123, 124, 186, 471, 486, 506, 615
Lead	123, 124, 125, 453, 471, 486, 506, 507, 615
Manganese	123, 124, 215, 471, 506, 615
Mercury	123, 124, 245, 471, 476, 486, 506
Molybdenum	615
Nickel	31, 33, 123, 125, 320, 453, 471, 615
Selenium	506
Zinc	31, 33, 123, 124, 453, 486, 506, 507
Other (general)	506, 615
Biocidal Contaminants	
Aldrin	186
DDT	690
Synthetic/Organic Contaminants	410, 666

TABLE 16 (continued)

Contaminant	Reference Number
Biological Contaminants	
Bacteria	38, 88, 347, 528
Clostridium botulinum	311
Clostridium perfringens	311
Coliforms	61, 293, 311, 472, 516
Coxsackie virus (A & B)	51, 88, 439
Escherichia coli	293
Fecal streptococci	293, 311, 528
Mycobacterium	223, 293, 311
Parasitic worms	88, 223
Polio virus	51, 53, 88, 411, 412, 439
Protozoa	223
Salmonella	88, 104, 223, 293, 311, 357
Shigella	88, 293, 311
Vibrio cholerae	88
Virus	38, 48, 49, 50, 54, 88, 185, 198, 200, 210, 223, 242, 259, 447, 488, 700
Other (general)	88, 280, 318, 690

chemical and biological contaminants of more direct public health concern.

The various activated sludge processes are all able to remove over 90 percent of the soluble BOD found in wastewater. Mitchell (453) recorded data on the removals achieved with activated sludge processes of these and other water quality contaminants during his study of the Los Angeles Hyperion Treatment Plant as shown in Table 17.

TABLE 17. ACTIVATED SLUDGE TREATMENT POLLUTANT REMOVALS, LOS ANGELES, CA (453)

	Treatment Influent (mg/ℓ)	Treatment Effluent (mg/ℓ)	Percent Removal
COD	315	31	90
BOD_5	165	9	95
Suspended Solids	103	9	91
Oil and Grease	28	0.5	98
Phenols	0.09	0.009	90
Ammonia nitrogen	20	9.6	52
Phosphorus	10.1	3.3	67
Cyanide	0.30	0.13	57

As can be seen from this table, relatively high removals of most water quality contaminants can be attained in a practical application over an extended period of time. These removal efficiencies are supported by other researchers and reviewers, including Noland and Birkbeck (485), Huang et al. (302), Rickert and Hunter (543), Lindstedt and Bennett (389), and Besik (62).

ELEMENTAL CONTAMINANTS

Although the activated sludge process efficiently removes biodegradable organic materials, only limited removal of soluble elemental contaminants from the wastewater stream can be achieved. The removal of elemental contaminants is governed by two basic mechanisms: (1) the precipitation of metal hydroxides; and (2) the adsorption of elemental contaminants by the activated sludge floc. In either case, the elemental contaminants removed will be contained in the sludge.

When suspended solids removals were in the 90 to 95 percent range, 90 percent of the aluminum, iron, mercury, lead, and zinc settled readily with the biofloc, according to Nomura and Young (486). Chromium (VI) and nickel median removals of 77 percent and 50 percent, respectively, were recorded under the same conditions. Morgan (462) stated that the removal percentages for chromium could vary, depending upon the treatment process used. Chromium exists in sewer systems in Cr (III) and Cr (VI); concentrations depend upon pH. Cr (III) is readily adsorbed on particles or precipitated as $Cr(OH)_3$. Chromium entering as Cr (VI) experiences a strongly reducing environment (little or no dissolved O_2) in sewers and treatment plants, and is thus reduced to Cr (III) and either precipitated or adsorbed. Aeration used in activated sludge or stabilization processes can cause the resolubilization by oxidation of trivalent to hexavalent chromium, with resulting effluent water degradation.

The association of trace metals with suspended solids during the activated sludge process was investigated by Chen and Hendricks (123) and Chen and Lockwood (124) at the Hyperion Treatment Plant. Their work confirms that many trace metals are associated with suspended solids, although the concentration of trace metals on particles does not appear to depend significantly on particle size. Rather, the removal of trace metals from the waste stream by activated sludge processes depends to a great extent upon the adsorptive capability of the activated flocs.

Mitchell (453) has recorded elemental removal efficiencies for the Hyperion Plant over a three-year period, as shown in Table 18.

TABLE 18 . REMOVALS OF TRACE METALS
BY ACTIVATED SLUDGE PROCESSES,
LOS ANGELES, CA (453)

Element	Influent (mg/ℓ)	Effluent (mg/ℓ)	Percent Removal
Copper	0.25	0.08	68
Zinc	0.42	0.23	46
Silver	0.019	0.012	37
Nickel	0.24	0.15	38
Lead	0.07	0.08	--
Arsenic	0.017	0.013	24
Cadmium	0.02	0.013	35
Chromium	0.37	0.013	96

These figures can be compared with the removal percentages assembled by the state of California as shown in Table 19. Clearly, the activated sludge process can reduce, but will not eliminate, trace metal concentrations in the municipal wastewater stream.

TABLE 19. PERCENT REMOVALS OF TRACE METALS BY THE ACTIVATED SLUDGE PROCESS (615)

Element	Average Percent Removal
Cadmium	56
Chromium	36
Copper	59
Iron	48
Lead	48
Manganese	22
Molybdenum	23
Nickel	22
Silver	71
Zinc	60

SYNTHETIC/ORGANIC CONTAMINANTS

Malaney et al. (410) in a study of the removal of possible carcinogenic organic compounds by activated sludge, concluded that no significant reduction was accomplished within normal detention times at the three treatment plants studied. Table 20 lists the possible carcinogens included in the analysis.

Recent work by Wachinski et al. (666) suggests that herbicide detoxification can be achieved with a pure oxygen-activated sludge treatment system that was determined to be both economical and ecologically safe. A proprietary strain of mutant microorganisms, PHENOBAC (developed by the Worne Biochemical Corp.), was utilized that was able to degrade halogenated phenols. Even with relatively high herbicide concentrations (1380 mg/ℓ), degradation of as much as 73 percent was accomplished after a 16-day aeration period using optimum proportions of required nutrients, microflora, and oxygen. According to the authors, this figure represents a conservative estimate of possible reductions, since testing was conducted at 18°C, while the optimum growth temperature for PHENOBAC is close to 30°C.

BIOLOGICAL CONTAMINANTS

Activated sludge followed by secondary sedimentation can remove over 90 percent of coliform or pathogenic bacteria that remain after primary sedimentation; other biological pathogens are removed to varying degrees. Nonetheless, even with 90 percent removal, appreciable amounts of pathogens remain present in the effluent.

TABLE 20. POSSIBLE CARCINOGENS INCLUDED IN THE ANALYSIS (410)

2,3 - Butylene oxide
B - Propiolactone
Thiourea
Ethycarbamate
2 - Thiouracil
4 - Ethoxyphenylurea
Benzidine
4,4' - Dihydroxy-a,b-diethylstilbene
2 - Naphthylamine
4,4' - Bis (dimethylamino) benzopheuone
p-Phenylazophenol
p-Phenylazoaniline
9,10 - Dimethylanthracene
1,2 - Benzanthracene
7 - Methyl-1,2-benzanthracene
9,10 - Dimethyl-1,2-benzathracene
1,2,5,6 - Dibenzanthracene
3/4 - Benzpyrene
1,2,4,5 - Dibenzpyrene
20 - Methylcholanthrene
2 - Nitrofluorene
2 - Fluoreneamino
N-2 - Fluorenylacetamide
7,9 - Dimethylbenz (c) acridine
7,10 - Dimethylbenz (c) acridine
Dibenz (a,h) acridine
Dibenz (a,j) acridine

Pathogens can either be removed by adsorption onto the sludge flocs or destroyed by the predatory activity of the zoogleal component. A review of the literature (280, 318) reveals discussion of general wastewater removal rates with little differentiation between removals and the biocidal properties of activated sludge.

Foster and Engelbrecht (223) provided a review of pathogen removal by activated sludge processes. Conclusions of this review are summarized in Table 21.

TABLE 21. REMOVAL OF PATHOGENS BY THE ACTIVATED SLUDGE PROCESS (223)

Pathogen	Percent Removal
Salmonella	96 to 99
Mycobacterium	Slight to 87
Amoebic cysts	No apparent removal
Helminth ova	No apparent removal
Virus	76 to 99

Ova of intestinal parasites are apparently unaffected by the activated sludge process; in fact, the literature indicates that activated sludge-mixed liquor provides an excellent hatching medium for eggs.

A review by Hunter and Kotalik (311) provided additional pathogen removal data, as summarized in Table 22.

TABLE 22. PERCENT REMOVALS OF BIOLOGICAL PATHOGENS BY THE ACTIVATED SLUDGE PROCESS (311)

Pathogen	Percent Removal
Coliform	90 to 99
Fecal streptococci	84 to 94
Shigella	90 to 99
Salmonella	70
Pseudomonas aeroginosa	99
Clostridium perfringens	90 to 99
Mycobacterium tuberculosis	66 to 88

A list of the species of protozoans, nematodes, and fungi that have been found in activated sludge effluent was also presented by the authors. Species recorded include:

Protozoa -

Amoeba sp.
Epistylis pliciatils
Euplotes patella
Loxophyllum helus
Oikomonas sp.
Pelodinium reniforme
Phyllomitus anylophagus
Trigonomonas compressar
Vorticella campanula

Nematodes -

Dorylaimus
Monhystera
Rhabdias

Fungi -

Alternaria
Aspergillus
Aureobasidium
Candida
Cryptococcus
Fusarium
Fusarium aquaeductum
Fusarium oxysporum
Fusarium roseum
Geotrichum
Hansenula
Kloeckera
Mucor
Penicillium
Rhodotorula
Saccharomyces
Torulopsis
Trichoderma
Trichosporon

The removal of viral contaminants by activated sludge has recently become the topic of considerable research. In general, viral removal of up to 90 percent has been observed after the activated sludge process. However, large variations in removal have been reported, probably because sampling was not temporally coordinated (242). Destruction by sewage microflora and virus adsorption to floc during the process are believed to be the main factors governing viral removal. Table 23 reports typical viral removals that can be expected from activated sludge treatment (88).

TABLE 23. VIRAL REMOVAL BY ACTIVATED SLUDGE TREATMENT (88)

Pathogen	Percent Removal
Enteroviruses	0 to 90 percent
Polio viruses	0 to 90 percent
Coxsackie viruses	0 to 50 percent
ECHO viruses	no apparent removal

Malina et al. (411) concluded from their research that viral inactivation by activated sludge is independent of the hydraulic detention time. Polio virus adsorption to sludge is almost immediate; the adsorbed virus particles are inactivated according to first order kinetics with a half-life in the range of 4 to 5 hr. Activated sludge utilizes aeration for optimum dispersion of the flocculant masses which, along with gases produced during microbial respiration, may entrain bacterial and viral pathogens in aerosols. The aerosols released are in the 5-μm diameter range, small enough to permit lung penetration of a substantial proportion of the particles.

SECONDARY TREATMENT: TRICKLING FILTER

INTRODUCTION

Trickling filters have been widely used for secondary biological treatment of municipal wastewater, and substantial literature is available on this process, as indicated on Table 24.

The trickling filter system generally consists of a tank open on both top and bottom. The tank is filled with a rock or plastic filter media having a high surface area to allow attachment of zoogleal slimes and void fraction for movement and diffusion of oxygen. Contaminant removal is accomplished through adsorption at the surface of the biological slimes covering the filter media. Following adsorption, the organics are utilized by the slimes for growth and energy. The trickling filter is followed by clarification to remove biological solids periodically flushed from the filter.

Trickling filter influent is usually from a primary treatment system. System outputs include effluent, sludge, and possible aerosols.

The literature generally refers to percent removal, with no distinction made between separation and degradation or destruction. As in the case of other secondary processes, the literature primarily addresses the general and biological contaminants and is sparse in the areas of elemental, synthetic, and biocidal contaminants.

WATER QUALITY PARAMETERS

The removal of BOD and suspended solids by trickling filters is reported to be from 65 to 95 percent, averaging about 85 percent (318, 615). The efficiency of trickling filtration decreases as temperatures fall below 20°C. Imhoff et al. (318) reported that a reduction of temperature from 20° to 10°C results in an efficiency loss of about 40 percent. Nickerson et al. (479) found that chemical addition ahead of primary clarifiers increases overall BOD and suspended solids removals in trickling filters. Lager and Smith (368) reported that no significant removal of total nitrogen or phosphorus occurred during the conventional trickling filter process. In

TABLE 24. LITERATURE REVIEWED PERTAINING TO TRICKLING FILTERS

Contaminant	Reference Number
Water Quality Parameters	
Ammonia	4, 20, 183, 368, 440, 653, 685
BOD	25, 131, 318, 368, 398, 450, 479, 490, 540, 615, 635, 653, 706
Chlorides	450, 685
COD	131, 258, 440, 450, 686
Nitrates	3, 4, 183, 368, 440, 685
Nitrites	183, 440, 685
Phosphates	32, 324, 368, 398, 440, 490, 685
Suspended solids	261, 318, 368, 479, 490, 615, 653, 706
Total organic carbon	25, 131, 311
Other (general)	205, 206, 280, 318
Elemental Contaminants	
Aluminum	398
Boron	685
Cadmium	615
Chromium	31, 540, 615
Copper	31, 540, 615
Germanium	655
Iron	615, 685
Lead	615, 685
Manganese	615, 685
Molybdenum	615

TABLE 24 (continued)

Contaminant	Reference Number
Nickel	31, 431, 540, 615
Zinc	31, 540, 615
Synthetic/Organic Contaminants	131, 311
Biological Contaminants	
Bacteria	38, 76, 88, 210, 254
Coliforms	254, 293, 472, 592
Coxsackie virus (A & B)	88
Escherichia coli	293
Fecal streptococci	592
Mycobacterium	223
Parasitic worms	88, 223, 311
Polio virus	88
Protozoa	223
Salmonella	88, 223, 357, 592
Shigella	88
Vibrio cholerae	88, 357
Virus	38, 49, 50, 51, 54, 88, 198, 224, 259, 592
Other (general)	88, 254, 318

low-rate filters, ammonia and nitrogenous organic compounds are usually oxidized to yield high proportions of nitrates and some nitrites in the effluent; high-rate trickling filter effluents are low in nitrates and nitrites due to limited system oxidation. Barth et al. (32) reported total phosphorus removals of up to 75 percent with direct dosing of aluminate to a trickling filter.

ELEMENTAL CONTAMINANTS

Removal of elemental contaminants by trickling filters is not well documented. A summary of the literature on metal removals is shown in Table 25.

TABLE 25. TRICKLING FILTER PROCESS REMOVAL OF TRACE METAL CONTAMINANTS (615)

Element	Average Percent Removal
Cadmium	5
Chromium	19
Copper	47
Iron	46
Lead	36
Manganese	16
Molybdenum	15
Nickel	20
Silver	48
Zinc	56

Trace metal removals by trickling filters are substantially lower than those achieved with the activated sludge process because there is less formation and sedimentation of trace metal complexes.

BIOLOGICAL CONTAMINANTS

Trickling filters do not effectively remove many biological pathogens. Table 26 illustrates reported removal efficiencies. Organisms are adsorbed into the zoogleal slime but due to similar surface charges and morphology, biocidal effects are variable (318).

TABLE 26. REMOVAL OF PATHOGENS BY TRICKLING FILTERS (223)

Pathogen Group	Efficiency
Salmonella	88 to 99.9
Mycobacterium	66 to 99
Amoebic Cysts	11 to 99.9
Helminth Ova	62 to 76
Virus	0 to 84

Literature by Foster and Engelbrecht (223) revealed that trickling filters are capable of reducing paratyphoid organisms by 84 to 99 percent. A review by Hunter and Kotalik (311) showed 99.7 percent removal of Schistosoma mansoni ova. These authors also concluded that trickling filter effluents can contribute a major portion of the free living nematode population found in receiving waters.

Improperly operated low-rate trickling filters can provide an excellent breeding area for insects, especially filter flies (psychoda) and springtails. Trickling filters cannot be depended upon to produce significant or consistent viral reductions. Foster and Engelbrecht (223) reported removals ranging from 0 to 84 percent. Berg (51) speculated that even when viruses are adsorbed, they may eventually be replaced by other substances and leach out of the filter slime as a result of an equilibrium effect.

SECONDARY TREATMENT: AERATED LAGOONS

INTRODUCTION

Aerated lagoons are aerobic or facultative ponds in which mechanical aeration is used to increase the rate at which oxygen is made available to facilitate biological stabilization. The aeration also provides mixing for suspension of microbial floc. The biological process does not include algae, and organic stabilization depends on the mixed liquor that develops within the pond. Literature surveyed concerning aerated lagoons is listed in Table 27.

WATER QUALITY PARAMETERS

BOD removal by aerated lagoons is a function of aeration period, temperature, and the nature of the wastewater. The aeration of a typical domestic wastewater for five days at 20°C provides about 85 percent BOD reduction; lowering the temperature to 10°C reduces the efficiency to approximately 65 percent (280).

BIOLOGICAL CONTAMINANTS

A discussion of the literature by Parker (503) revealed that coliform reductions in the range of 80 to 99 percent can be achieved with optimum detention time. This is supported by the experiments of Carpenter et al. (105), who reported that coliform organisms are efficiently removed by the use of aerated lagoons. Klock (357) stated that the coliform survival rate in lagoons is a function of the oxidation-reduction potential and temperature.

Berg (49) discussed the removal of viruses by stabilization ponds, concluding that virus removal can be expected to be erratic.

TABLE 27. LITERATURE REVIEWED PERTAINING TO AERATED LAGOONS

Contaminant	Reference Number
Water Quality Parameters	
Ammonia	233
BOD	47, 252, 280, 332, 386, 654
COD	47, 143, 252
Nitrates	233
Nitrites	233
Phosphates	173, 386
Suspended solids	252, 368
Other (general)	206, 280, 318
Biological Contaminants	
Bacteria	105, 357
Coliforms	105, 293, 357, 503
Escherichia coli	503
Fecal streptococci	293, 503
Salmonella	293
Virus	49, 105, 200

SECONDARY TREATMENT: PONDING

INTRODUCTION

An oxidation or facultative pond is generally a shallow earthen basin designed to promote a symbiotic existence between algae and bacteria (368). Algal photosynthesis and surface reaction maintain aerobic conditions in the photic region, while anaerobic bacteria flourish in the aphotic zone. Ponds are normally operated in series and are sometimes used for "polishing" effluent from conventional secondary processes.

Influent to a ponding system may be raw sanitary waste, primary effluent, or secondary effluent. Pond effluent can enter the environment by direct discharge or by seepage to the groundwater. Literature reviewed concerning ponding is listed in Table 28; as can be seen from this review, only a limited amount of research has examined the removal of contaminants by the ponding process.

WATER QUALITY PARAMETERS

Oxidation pond removal efficiencies for suspended solids and BOD can vary widely and may even reach negative values (368). Removal ranges of 60 to 50 percent have been reported for suspended solids, and of 70 to 10 percent for BOD_5. This variation occurs because most influent BOD is converted into suspended algal mass. This mass exerts a BOD demand and provides suspended solids that may be carried out in the effluent.

Bacterial decomposition and algal growth are both retarded, reducing removal efficiency of the ponding process, by reduced temperatures (280).

SYNTHETIC/ORGANIC CONTAMINANTS

The removal of trisodium nitrilotriacetate (NTA) by ponding has been investigated by Klein (356). He found that after a two-month acclimation period, steady-state removal was in excess of 90 percent, with influent concentrations in the range of 30 mg/ℓ.

TABLE 28. LITERATURE REVIEWED PERTAINING TO PONDING

Contaminant	Reference Number
Water Quality Parameters	
BOD	172, 280, 368
COD	685
hlorides	222
Fluorides	222
Nitrates	122, 172, 195, 230, 663
Phosphates	172, 195, 221, 230, 685, 693
Suspended solids	206, 368
Other (general)	206, 280, 368
Synthetic/Organic Contaminants	356
Biological Contaminants	
Bacteria	413, 592
Coliforms	417, 480, 592
Escherichia coli	418, 500
Fecal streptococci	417, 418, 592
Salmonella	337
Virus	49, 50, 51, 592

BIOLOGICAL CONTAMINANTS

Kampelmacher and Jansen (337) found that removal of salmonella by oxidation ponds was not inferior to removal achieved by conventional treatment plants. Species of the coliform group, although reduced by ponding, are not effectively eliminated according to Parhad and Rao (500). Slanetz et al. (592), however, reported that if two ponds were operated in a series at temperatures of 17° to 26°C, the die-off rate of coliform, fecal coliforms, and fecal streptococci ranged from 95 to 99 percent. During winter when temperatures were in the 1° to 10°C range, the die-off rate was 46 times lower. Berg (51) states that virus removal by ponding is erratic, ranging from 0 to 96 percent; virus recovery decreased as the effluent passed through a series of maturation lagoons.

SECTION 5

TERTIARY TREATMENT: FILTRATION

INTRODUCTION

Inability of gravity sedimentation in secondary clarifiers to remove small particles (and associated public health impairing contaminants) is a limitation of BOD and suspended solids removal by conventional wastewater treatment. Filtration as a tertiary process upgrades treatment performance by removing a portion of the unsettled suspended solids from secondary effluents. In addition, filtration often precedes other tertiary processes such as adsorption and ion exchange since the presence of suspended solids interferes with the operation of these processes.

Filtration of wastewater to reduce the suspended solids concentration is accomplished by passage through a bed of granular particles. Single, dual, or mixed media beds may be used, composed of anthracite coal, granite, sand, and/or gravel (280). Suspended solids are removed by a variety of mechanisms: straining, impingement, settling, and adhesion. The treatment efficiency of the process is influenced by:

- The concentration and characteristics of the wastewater solids (particle-size distribution, surface characteristics, organic versus inorganic, etc.);
- The characteristics of the filter media and filtering aids used (particle-size distribution, surface characteristics, etc.);
- The design and operation of the filter.

Since wastewater flow rate and solids content are variable, and processes upstream of filtration may vary in performance, the efficiency of filtration may also be expected to vary. For this reason, values presented in the following discussion should be considered as merely indicative of the range of achievable removals.

TABLE 29. LITERATURE REVIEWED PERTAINING TO FILTRATION

Contaminant	Reference Number
Water Quality Parameters	
Ammonia	21, 287, 438, 615, 706
BOD	64, 166, 348, 377, 438, 611, 615, 649, 706
COD	64, 377, 438, 611, 615
Chlorides	62, 438, 506, 611, 638
Cyanides	506
Fluorides	506
Nitrates	202, 438, 611, 638
Oil & grease	506
Phosphates	59, 64, 166, 335, 438, 615, 649
Suspended solids	19, 59, 233, 280, 335, 377, 615, 638, 649, 670, 706
Total dissolved solids	506
Total organic carbon	62, 64, 615
Other (general)	203, 280, 318
Elemental Contaminants	
Arsenic	506, 576
Barium	506
Boron	506
Cadmium	18, 19, 391, 506, 507, 714
Chromium	18, 19, 391, 506, 714
Copper	18, 19, 348, 506, 507, 611, 714
Iron	18, 506, 611, 714

TABLE 29 (continued)

Contaminant	Reference Number
Lead	506, 507, 714
Manganese	18, 325, 506, 611, 714
Mercury	506
Nickel	18, 506, 507
Selenium	18, 391, 506
Zinc	18, 348, 506, 507, 611, 714
Other	18, 506
Synthetic/Organic Contaminants	506, 691
Biological Contaminants	
Bacteria	88, 176
Coliforms	55, 243, 305, 313
Coxsackie virus (A & B)	49, 88
ECHO virus	560
Hepatitis virus	243
Parasitic worms	88, 678
Polio virus	56, 81, 88, 118, 547, 560, 601
Salmonella	88
Shigella	88
Vibrio cholerae	88, 219
Virus	14, 49, 50, 80, 82, 88, 139, 176, 233, 242, 259, 305, 487, 574, 608
Other	81, 88

WATER QUALITY PARAMETERS

In general, the best effluent quality achievable by plain filtration of secondary effluent is about 5 to 10 mg/ℓ for suspended solids and BOD. The suspended solids content of secondary effluent was reduced to 5 mg with both rapid sand and mixed media filters, employed respectively at a treatment and a pilot plant. Complete removal, however, could not be effected. If further reduction is desired, chemical coagulation must precede filtration (233, 280).

Two studies (615) indicated that essentially complete suspended solids removal was accomplished at both a pilot facility and a major treatment plant when filtration was preceded by chemical treatment of secondary effluent. Filter effluent contained 3 mg/ℓ BOD and 25 mg/ℓ COD. Chemical clarifier effluent contained 0.7 mg/ℓ total phosphorus; after filtration, this phosphorus content was reduced to 0.1 mg/ℓ . At the pilot facility, the filter effluent contained 17.6 mg/ℓ COD, 9 mg/ℓ total organic carbon (TOC), and no detectable phosphorus, compared with filter influent concentrations of 18.1 mg/ℓ COD, 8.6 mg/ℓ TOC, and 0.4 mg/ℓ phosphorus.

When chemically treated secondary effluent was applied to rapid-sand filters, generally 20 to 60 percent of applied BOD was removed, 30 to 70 percent of phosphate, and 40 to 80 percent of the suspended solids (649). These values were obtained using a variety of influent concentrations and chemical dosages. The results of 30 days continuous operation of a pilot plant practicing filtration preceded by chemical treatment of secondary effluent are summarized in Table 30 (615).

TABLE 30. RESULTS OF ONTARIO, CANADA PILOT PLANT STUDY INVOLVING FILTRATION PRECEDED BY CHEMICAL TREATMENT OF SECONDARY EFFLUENT (615)

Quality Parameter	Raw Wastewater	Secondary Effluent	Filter Effluent
Total Organic Carbon (mg/ℓ)	110-165	14-28	4.5-7.5
BOD_5 (mg/ℓ)	230-400	5-14	2.0-3.0
PO_4 (as PO_4)	9-21	1.3-3.5	0.4-1.0
Total Nitrogen N	27-51	25-37	20-35
Ammonia N	17-29	21-29	18-29
Suspended Solids (mg/ℓ)	148-268	13-37	3-12

Results of a pilot plant study at Cleveland, Ohio - where chemical coagulation and settling of raw wastewater was followed by filtration and granular carbon adsorption - were reviewed by Culp and Shuckrow (151). On the basis of the data provided, removals attributable to chemical treatment and filtration can be calculated to be 66 percent of the applied BOD and 77 percent of the applied COD. A second pilot plant study reviewed by the authors involved the same treatment scheme. Removals at this plant due to combined chemical treatment and filtration can be inferred to range from 77 to 84 percent.

While studying soil filtration, de Vries (166) applied primary effluent to a sand filter and obtained BOD and phosphate removals of nearly 100 percent. Phosphate removals were attributed to the natural coatings of Fe_2O_3 and Al_2O_3 on sand grains.

ELEMENTAL CONTAMINANTS

After chemical treatment, filtration to remove residual particulate matter may provide some additional removal of elemental contaminants. Elemental contaminant removals achieved by filtration depend primarily upon the extent of suspended solids removals, with which the various trace elements are associated. Table 31, compiled from the literature by Argo and Culp (18) gives results for sand filtration of some municipal and industrial wastes.

TABLE 31. HEAVY METAL REMOVAL BY SAND FILTRATION FOLLOWING LIME COAGULATION (18)

Metal	Concentration Before Filt.	Concentration After Treat.	pH	% Removal By Filt.
Cd	Trace to 0.00075 mg/ℓ	 0.00070	8.1 7.6	95 6.6
Cr^{+6}	0.0503	0.049	7.6	2.6
Cr^{+3}	2.7	0.63	8.7	77
Cu	0.79 -	0.32 .5	9.5	59.5
Fe	-	0.1 1.2 Organic	10.8 10.5	
Mn	-	0.1 1.1 Organic	10.8 10.5	
Ni	0.08	0.1 0.5	8.7 9.5	
Se	.0103	0.00932	11	9.5

TABLE 31 (continued)

Metal	Concentration Before Filt.	Concentration After Treat.	pH	% Removal By Filt.
Ag	0.00164	0.00145	11	11.6
Zn	0.97	0.23 2.5	8.7 9.5	76.3

Patterson (506) cites evidence from pilot plant studies that little or no additional removal of arsenic was afforded by filtration of chemically treated municipal wastewater.

BIOLOGICAL CONTAMINANTS

Several investigations have been reported concerning the removal of viruses by sand filtration. It has been shown that insignificant virus removal is achieved by rapid filtration through clean sand (14, 49). However, virus removal efficiency will be increased by impregnation of the filter medium with coagulated floc, the presence of organic matter trapped in the sand, chemical flocculation prior to filtration, or a decrease in the filtration rate. The addition of iron salts prior to filtration has resulted in significantly higher coliform reductions, as discussed by Hunter et al. (313). Similarly, Robeck et al. (547) noted that if a low dose of alum was fed to a rapid coal and sand filter just ahead of filtration, more than 98 percent of polio virus Type I could be removed. If the dosage was increased and conventional flocculators and settling were used, removal was increased to over 99 percent. The authors also noted a general trend toward better removal of polio virus I with slower filtration rates although their data were generally erratic. At slow sand filter rates (0.6 to 1.2 $\ell/\text{min}/\text{m}^2$), removal ranged from 50 percent to about 98 percent. At rapid filtration rates (38 to 76 $\ell/\text{min}/\text{m}^2$), virus removals ranged from about 10 to 70 percent. Similarly, research examined by Berg (49) showed that filtration through sand at 7.5 $\ell/\text{min}/\text{m}^2$ removed 99 percent of the coxsackie virus A5 while filtration at 75 $\ell/\text{min}/\text{m}^2$ removed only about 10 percent of the virus.

Brown et al. (81) reported 70 to 90 percent removals of low concentrations of either bacteriophage T2 or polio virus Type I by filtration through uncoated diatomaceous earth. However, no significant virus removals by uncoated diatomaceous earth were achieved in a laboratory study by Amirhor and Engelbrecht (14). With polyelectrolyte-coated filter media, removals greater than 99 percent were consistently achieved.

In laboratory tests by Berg et al. (56), from 82 to greater than 99.8 percent of polio virus I was removed from chemically treated effluent by rapid sand filtration. The results of these tests are given in Table 32 .

TABLE 32 . REMOVAL OF POLIOVIRUS I FROM
$Ca(OH)_2$ FLOCCULATED EFFLUENT BY RAPID
SAND FILTRATION*
AS MEASURED BY MEMBRANE FILTER RECOVERY OF VIRUS (56)

Test No.	Virus Concentration pfu/ℓ Before Sand Filtration †	Virus Concentration pfu/ℓ After Sand Filtration	Virus Removal percent
1	2,200	397	82.0
2	15,912	750	95.3
3	1,940	<4.6	>99.8
4	505	12.5	97.5
5	47	2.8	94.0

* Filtration rate 2.25 gpm/sq ft through 8 in of sand.

† Virus concentration in flocculated effluent just prior to sand filtration.

Laboratory experiments on the removal of nematodes by rapid sand filtration were conducted by Wei et al. (678). Removal efficiency was about 96 percent when all the nematodes in the influent were dead or nonmotile. However, most motile nematodes were able to penetrate the filter bed.

Sand filtration may also provide some removal of amoebic cysts and ascaris eggs, according to a literature survey by Bryan (88). He did not indicate the levels of removal afforded.

TERTIARY TREATMENT: ADSORPTION

INTRODUCTION

Adsorption refers to the removal from water or wastewater streams of dissolved contaminants by their attraction to and accumulation on the surface of an adsorbent substance. Activated carbon is the most widely used adsorbent in municipal wastewater treatment to remove trace organics. Adsorption using activated carbon is utilized as a tertiary treatment step, usually following sand or multimedia filtration.

Carbon adsorption systems generally utilize granular or powdered activated carbon packed in a column or forming a filter bed through which wastewater is passed. Three consecutive steps occur in the adsorption of wastewater contaminants by activated carbon: (1) the film diffusion phenomenon, or the transport of the adsorbate through a surface film surrounding the activated carbon; (2) the diffusion of the adsorbate within the pores of the activated carbon; and (3) adsorption on the interior surfaces of the activated carbon.

Carbon adsorption of contaminants has been the topic of many research projects as shown in the literature surveyed for this report, tabulated in Table 33 .

WATER QUALITY PARAMETERS

Adsorption is most effective for removing refractory and other organics from wastewater. This is especially important when effluents of exceptional quality are required (e.g., for groundwater recharge or other reuse applications). Adsorption can be used either as a polishing step or as the major treatment process (676). Rizzo and Schade (544) and Zanitsch and Morand (712) reported that carbon columns alone were capable of about 85 percent removal of BOD from wastewaters entering the columns. Bishop et al. (64, 68) and Zanitsch and Morand (712) reported 75 to 80 percent TOC removals under the same conditions. Weber et al. (676) found that a treatment system composed of primary settling, ferric chloride coagulation, and carbon adsorption could remove up to 97 percent of the influent BOD.

TABLE 33. LITERATURE REVIEWED PERTAINING TO ADSORPTION

Contaminant	Reference Number
Water Quality Parameters	
Ammonia	61, 64, 65, 103, 438
BOD	61, 64, 65, 84, 92, 103, 151, 167 172, 261, 277, 368, 438, 544, 676, 712
COD	64, 65, 84, 103, 151, 174, 261, 336, 438, 544, 707
Chlorides	62, 438, 494, 598
Nitrates	61, 64, 65, 172, 676
Oil and grease	249
Phosphates	13, 61, 64, 65, 92, 103, 172, 232, 438, 676
Suspended solids	19, 61, 64, 65, 92, 103, 172, 261, 336, 438, 676
Total dissolved solids	103, 233, 707
Total organic carbon	61, 62, 64, 65, 68, 167, 277, 518, 544, 587, 676, 712
Other (general)	280, 318
Elemental Contaminants	
Aluminum	644
Arsenic	419, 644
Barium	419, 644
Boron	538, 644
Cadmium	18, 19, 315, 370, 372, 419, 549, 644
Chromium	18, 19, 303, 315, 370, 372, 419, 549, 644

TABLE 33 (continued)

Contaminant	Reference Number
Elemental Contaminants	
Cobalt	370, 644
Copper	19, 316, 549, 644
Iron	316, 370, 644
Lead	370, 419, 644
Manganese	370, 419, 644
Mercury	370, 394, 419
Nickel	370, 419, 644
Selenium	18, 506, 644
Zinc	316, 419, 644
Other (general)	18, 372
Biocidal Contaminants	
Chlorinated hydrocarbons	373, 570
Dieldrin	278
Herbicides	370, 272, 570
Other (general)	278, 322, 373, 615
Synthetic/Organic Contaminants	67, 167, 233, 249, 278, 322, 332, 373, 422, 541, 599, 615, 674, 691
Biological Contaminants	
Bacteria	154
Coliforms	61
Escherichia coli	140
Polio virus	118, 240, 241, 601, 609
Virus	133, 140, 242, 259, 488, 608, 609

There is some disagreement among researchers regarding the ability of activated carbon to remove nitrogen species from wastewaters. Bishop et al. (64) reported that carbon adsorption had little effect on nitrogen concentrations. On the other hand, Weber et al. (676) found that their primary settling ferric chloride coagulation, carbon adsorption system removed 95 percent of the influent nitrate; the reduction was attributed partly to biological populations growing in the columns.

Weber et al. (676) reported that their pilot system also removed 90 percent of the phosphate in the influent wastewater. However, much of the research on phosphate adsorption has been concerned with adsorbents other than carbon. Gangoli and Thodos (232) reported that both F1 Alumina and fly ash were capable of removing up to 99 percent of influent phosphate levels. Ames and Dean (13) demonstrated that an alumina column could treat up to 400 column volumes before reaching the 10 percent phosphorus breakthrough level.

Since color is contributed largely by organic compounds in water, high color removal levels with adsorption should be possible. Zanitsch and Morand (712) demonstrated 90 percent color removal. They also noted an 86 percent suspended solids removal, presumably by filtration.

ELEMENTAL CONTAMINANTS

Not a great deal of literature is available on the removal of elemental contaminants by carbon adsorption. Such systems are not specifically designed to remove ionic elemental contaminants, but some elementals are incidentally removed. When the metallic contaminants are in an organometallic complex, carbon adsorption columns can remove specific species. Literature from several sources (315, 372) reveals that high removals (95 percent) of cadmium and hexavalent chromium by carbon adsorption are possible. Huang and Wu (303) found that the efficiency of chromium removal increases with decreasing chromium concentration. While the mechanism of removal is not well understood, Roersma et al. (549) were able to describe the reduction of Cr^{+6} to Cr^{+3}. The Cr^{+6} is adsorbed within the pores of the activated carbon which, in turn, is slowly oxidized to CO_2, reducing the chromium ion.

Activated carbon treatment of a secondary-treated municipal wastewater was found to reduce selenium from 9.32 to 5.85 ppb in a study cited by Patterson (506). This represents a 37 percent removal efficiency. Patterson cited a second study in which the selenium removal efficiencies of several advanced wastewater treatment processes were determined in the treatment of secondary effluent containing 2.3 ppb selenium. While sand filtration alone reduced the effluent concentration by 9.5 percent, sand filtration followed by activated carbon treatment provided a cumulative removal of 43.2 percent of the selenium from the secondary effluent.

Logsden and Symons (394) researched the removal of mercury by carbon adsorption and found that powdered carbon, in a jar test, would adsorb both inorganic and methyl forms in excess of 70 percent.

BIOCIDAL CONTAMINANTS

Carbon adsorption is widely applied to remove organic or metal-organic biocides. The removal of insecticides and pesticides has been reviewed by Hager and Flentje (278). Dieldrin, lindane, parathion, and 2, 4, 5-T ester were reduced below the detectable limit of 0.01 ppb with influent concentrations of 3.6 to 11.4 ppb. Influent concentrations of 3, 5 dinitro-o-cresol of 30 to 180 ppb were reduced to less than 1 ppb by carbon adsorption. It was concluded that granular carbon beds will provide a margin of safety for treatment of water containing varying insecticide or pesticide residues.

Activated carbon removals of several pesticides and PCB's are well illustrated by results of laboratory studies cited by the California State Water Resources Control Board (615), as shown in Table 34 . A variety of pesticides were experimentally added to distilled water and passed through carbon filters to test removal efficiencies. Schwarz (570) investigated the adsorption of isoprophyl N-(3-chlorophenyl) carbamate (CIPC) onto activated carbon, concluding that powdered activated carbon readily adsorbs CIPC from aqueous solution. The adsorption of CIPC on activated carbon was independent of the pH in the range of 5 to 9.

Grover and Smith (272) studied adsorption onto activated carbon of the acid and dimethylamine forms of 2, 4-D, and dicarbamate. A strong adsorption effect was noted on both the acidic and salt forms of the compounds. This effect was expected to increase at low pH values.

On the basis of the literature which has been reviewed, it can be concluded that activated carbon adsorption is effective in the removal of some biocidal contaminants; however, further investigation of this process will be useful.

SYNTHETIC/ORGANIC CONTAMINANTS

As was mentioned earlier, activated carbon is most effective at removing organic contaminants from aqueous solutions. It is particularly effective at removing organics of low water solubility, as are many synthetic organic compounds. In general, carbon adsorption following secondary treatment is capable of

TABLE 34. REMOVAL OF SPECIFIC TOXIC MATERIALS BY CARBON ADSORPTION (615)

Carbon Dosage (mg/ℓ)	Residual (ppb)								
	Aldrin	Endrin	Dieldrin	DDT	DDD	DDE	Toxaphene	Arochlor 1242	Arochlor 1254
Control	48	62	19	41	56	38	155	45	49
1.0	--	--	--	41	--	34	147	--	--
2.0	26	15	6.3	--	6.9	--	80	7.3	37
2.5	--	--	--	21	--	29	--	--	--
5.0	15	3.4	2.4	3.7	3.7	12	31	1.6	17
10.0	12	1.5	1.1	--	2.2	--	2.7	1.1	4.2
12.5	--	--	--	<1	--	3.3	--	--	--
25.0	6.3	0.56	--	--	0.45	1.1	--	--	1.6
50.0	4.4	0.22	--	--	0.35	0.9	--	--	1.2

producing an effluent with from 1 to 7 mg/ℓ of organic carbon (233).

Bishop et al. (68) found that carbon adsorption was least effective in removing highly polar, highly soluble organic species. DeWalle and Chian (167) found that removal was a function, at least in part, of the molecular weight of the synthetic organic contaminant: low (<100) and high (>50,000) molecular weight compounds are poorly adsorbed. The major fraction removed by adsorption falls into a 100 to 10,000 molecular weight range.

Studies of the adsorption of 93 petrochemicals by Giusti et al. (249) confirmed the results of Bishop et al. (68) and DeWalle and Chian (167); adsorption is largely a function of molecular weight, polarity, solubility, and branching. Ability to function substantially affected solubility and polarity. The relative amenabilities to carbon adsorption of straight chain molecules of fewer than four carbon atoms were as follows: >aldehydes >esters >ketones >alcohols >glycols. For larger molecules, the alcohols moved ahead of the esters.

Much of the research done on the adsorption of synthetic organic compounds from wastewater has been concerned with determining mechanisms of adsorption and optimum removal conditions. The only general removal efficiency studies available report removals in terms of total organic carbon with little or no effort made to differentiate the organic compounds involved. Based on these results, adsorption can reduce the levels of synthetic organic compounds in a typical domestic wastewater by 75 to 85 percent. If a particular type or types of organic compounds predominate in a wastewater, these removals must be adjusted to reflect the effect of compound character on the adsorption process.

BIOLOGICAL CONTAMINANTS

With the exception of enteroviruses, no information was found on the adsorption of biological contaminants, although incidental removal of other organisms would be expected by filtering action. Adsorption brings about simple removal of viruses from wastewater rather than inactivation or destruction (140, 242). Consequently, viable viruses could be reintroduced to wastewater should desorption of viruses adsorbed to activated carbon occur.

Columns of granular activated carbon removed between 18 and 40 percent of Type I polio virus from secondary effluent in studies by Sproul et al. (609). This research and research by Gerba et al. (241) using Type I polio virus indicate that adsorption is inversely related to the concentration of organic matter in the wastewater. The organics and the virus compete for adsorption sites; consequently, desorption of virus can occur as adsorption of organic matter continues, or if the concentration of organics is increased. Several authors have thus concluded that the process is not dependable for producing virus-free effluents (133, 242, 609).

The level of removal actually attained is closely related to the type of treatment that precedes adsorption. For example, reducing the concentration of soluble organics in wastewater by lime coagulation increased polio-virus removal in studies by Gerba et al. (240, 241). In addition, the degree of virus adsorption from lime-treated wastewater exceeded that from filtered wastewater. These investigations also showed that poliovirus removal from wastewater effluents is greatly improved by maintaining a pH value in the range of 3.5 to 4.5. It was found that virus adsorbed at low pH could become desorbed by a rise in pH.

TERTIARY TREATMENT: CHEMICAL TREATMENT

INTRODUCTION

The purpose of chemical treatment is to coagulate suspended solids and cause the precipitation of phosphate and various trace metals. Chemical coagulation of secondary effluents may be accomplished by the addition of lime, alum, polymers, or iron salts, and involves three operations: (1) injection and rapid mixing of the coagulants to neutralize the predominantly negative charges on suspended matter; (2) gentle stirring to promote agglomeration of the coagulated particles into large, settleable floc; and (3) sedimentation to provide gravity separation of the flocculated material from the wastewater. The settled material is disposed to a sludge-handling system. As indicated by Table 35, a great deal of information is available concerning the removal of various public health impairing contaminants by chemical treatment processes.

WATER QUALITY PARAMETERS

Culp and Shuckrow (151) investigated chemical treatment of raw wastewater with lime and found that removals of 95 to 98 percent phosphorus and 99 percent suspended solids can be achieved with chemical clarification followed by carbon adsorption. The treatment of municipal wastewater with alum precipitation as studied by Shuckrow et al. (582) resulted in removal efficiencies of 85 percent for COD and 83 percent for total organic carbon.

The removal of BOD, suspended solids, and phosphorus as reviewed by Lager and Smith (368) is summarized in Table 36 Removals from secondary effluents of the magnitudes listed obviously provide a high quality effluent.

TABLE 35. LITERATURE REVIEWED PERTAINING TO CHEMICAL TREATMENT

Contaminant	Reference Number
Water Quality Parameters	
Ammonia	21, 65, 150, 155, 204, 301, 310, 438, 440, 613, 700
BOD	64, 69, 150, 151, 155, 178, 261, 310, 368, 387, 389, 397, 398, 432, 438, 479, 489, 490, 569, 622, 646, 649, 651, 676, 700
Chlorides	438
COD	64, 65, 69, 134, 150, 151, 155, 174, 261, 438, 582
Cyanides	58, 453, 477
Fluorides	215, 613
Nitrates	64, 65, 69, 134, 150, 151, 155, 174, 261, 438
Nitrites	155, 204, 440, 517, 613, 700
Phosphates	6, 10, 32, 59, 64, 65, 69, 150, 151, 155, 156, 160, 172, 173, 195, 215, 218, 248, 269, 301, 310, 331, 354, 367, 368, 376, 387, 389, 398, 426, 431, 438, 440, 489, 490, 495, 497, 567, 569, 590, 607, 613, 622, 625, 649, 651, 675, 693, 700, 707, 710
Suspended solids	18, 59, 64, 65, 134, 151, 175, 261, 301, 349, 368, 389, 397, 443, 479, 489, 490, 569, 582, 623, 649, 651, 686, 700
Total dissolved solids	150, 569, 613
Total organic carbon	64, 65, 69, 155, 582, 676
Other (general)	203, 205, 206, 280, 318

TABLE 35 (continued)

Contaminant	Reference Number
Elemental Contaminants	
Aluminum	178, 398, 495, 644
Antimony	18
Arsenic	18, 419, 453, 506, 615, 644
Barium	18, 419, 506, 615, 644
Boron	644
Cadmium	18, 19, 58, 391, 419, 453, 525, 615, 644, 696, 697, 698
Chromium	18, 19, 106, 391, 419, 615, 644, 696, 697, 698
Cobalt	644
Copper	18, 19, 58, 106, 419, 453, 615, 644, 696, 697, 698
Iron	18, 178, 376, 615, 644, 696, 698
Lead	419, 453, 615, 644, 696, 698
Manganese	18, 178, 215, 419, 615, 644, 696, 697, 698
Mercury	18, 379, 394, 419, 453, 513, 615, 631, 644, 696, 697, 698
Molybdenum	18, 615
Nickel	18, 419, 453, 615, 644, 696, 697
Selenium	391, 615, 644
Uranium	18, 615
Zinc	18, 58, 419, 453, 615, 644, 696, 697, 698
Other (general)	18, 372, 389, 615, 698

TABLE 35 (continued)

Contaminant	Reference Number
Synthetic/Organic Contaminants	5, 691
Biological Contaminants	
Adeno virus	705
Bacteria	38, 176, 179, 338, 389, 407, 649
Coliforms	55, 155, 243, 389, 443, 649
Coxsackie virus (A&B)	51, 705
CHO virus	705
Escherichia coli	433, 561, 643
Fecal streptococci	389
Hepatitis	241, 705
Parasitic worms	407
Polio virus	56, 86, 118, 262, 601, 609, 643, 700, 705
Protozoa	407
Salmonella	338
Virus	38, 48, 50, 51, 86, 116, 117, 132, 133, 138, 154, 155, 176, 198, 199, 242, 416, 487, 504, 510, 575, 602, 608, 609, 700, 701

TABLE 36. REMOVALS ACHIEVED BY CHEMICAL CLARIFICATION (368)

Chemical	BOD Removal (Percent)	Suspended Solids Removal (Percent)
Lime pH 11.5	80	90
Ferric Chloride 170 mg/ℓ dose	80	95
Ferric Chloride 80-100 mg/ℓ dose	75	--

Phosphate removal by chemical precipitation (lime) has received considerable attention in the literature in recent years. The research on phosphate removal in general indicates that lime clarification usually provides removal efficiencies greater than 90 percent. This is supported by the work of Davis (160), Sturm and Hatch (625), Johnson (331), and Bernhardt et al. (59).

ELEMENTAL CONTAMINANTS

The precipitation of metal hydroxides from solution is governed by the pH and the concentration of the metal ion in solution. Since many of the trace metals form insoluble hydroxides near pH 11, lime coagulation results in a reduction of these metal concentrations. Table 37 (615) summarizes the effects of lime coagulation on a number of heavy metals. Some of the data were collected on industrial metal wastes characterized by metal ion concentrations a great deal higher than would occur in any municipal plant influent. The data are included here as examples of possible metal reductions, since such figures from chemical coagulation of municipal wastewaters are scarce.

As can be seen from these figures, arsenic, molybdenum, and selenium had relatively poor removal rates,and the potential removal of mercury was estimated to be low. Only 11 percent of hexavalent chromium was removed, although the trivalent form was reduced more than 99.9 percent. Most other metals tested were very effectively reduced at high pH. Lower removals of these same metals (usually less than 50 percent) can be achieved with alum coagulation at near neutral pH values (615), a fact that illustrates the dependence of precipitation on pH.

TABLE 37 . REMOVAL OF ELEMENTAL CONTAMINANTS BY LIME COAGULATION (615)

Metal	Concentration Before Treatment mg/ℓ	Concentration After Treatment mg/ℓ	Percent Removal
Antimony[a]	--	--	90
Arsenic[a]	--	--	<10
	23	23	0
Barium[a]	--	1.3 (sol)[b]	
Bismuth[a]	--	.0002 (sol)	
Cadmium	Trace	--	Abt. 50
	0.0137	0.00075	94.5
Chromium (+6)	0.56	0.050	11
Chromium (+3)	7,400	2.7	99.9+
	15	0.4	97
Copper	15,700	0.79	99.9+
	7	1	86
	7	.05	93
	302	Trace	99+
	15	0.6	97
Gold[a]	--	<.001 (sol)	90+
Iron	13	2.4	82
	17	0.1	99+
	2.0	1.2[c]	40
Lead[a]	--	<.001 (sol)[b]	90+
	15	0.5	97
Manganese	2.3	<0.1	96
	2.0	1.1[c]	45
	21.0	0.05	95
Mercury[a]	--	Oxide Soluble	<10
Molybdenum	Trace	--	Abt. 10
	11	9	18

TABLE 37 (continued)

Metal	Concentration Before Treatment mg/ℓ	Concentration After Treatment mg/ℓ	% Removal
Nickel	160	0.08	99.9+
	5	0.5	90
	5	0.5	90
	100	1.5	99
	16	1.4	91
Selenium	0.0123	0.0103	16.2
Silver	0.0546	0.0164	97
Telurium[a,d]	--	(<0.001?)	(90+)
Titanium[a,d]	--	(<0.001?)	(90+)
Uranium[e]	--	?	?
Zinc	17	0.3	98
	--	.007 (sol)	90+

[a] The potential removal of these metals was estimated from solubility data.

[b] Barium and lead reductions and solubilities are based upon the carbonate.

[c] These data were from experiments using iron and manganese in the organic form.

[d] Titanium and telurium solubility and stability data made the potential reduction estimate unsure.

[e] Uranium forms complexes with carbonate ion. Quantitative data were unavailable to allow determination of this effect.

Pilot plant studies of municipal wastewater containing 5 mg/ℓ arsenic cited by Patterson (506) suggest that chemical treatment can provide efficient removal of this element. Ferric sulfate at 45 mg/ℓ Fe and pH 6.0 removed 90 percent of the arsenic; lime at 600 mg/ℓ and pH 11.5 removed 73 percent. In similar studies cited by Patterson, barium removals of 97 percent were obtained when municipal wastewater dosed with 5 mg/ℓ barium was treated with 45 mg/ℓ Fe at pH 6.0. Lime at 600 mg/ℓ and pH 11.5 resulted in 80 percent removal. The removal of cadmium from waters by sorption onto hydrous oxides of solid metals such as manganese and iron was investigated by Posselt and Weber (525). They concluded that sorptive uptake of cadmium on such materials would constitute a method easily adaptable to present treatment technology.

BIOLOGICAL CONTAMINANTS

Chemical treatment can be used to reduce or remove many biological pathogens present in municipal wastewater. Lindstedt and Bennett (389) evaluated the effectiveness of lime clarification in reducing bacterial concentrations, finding that treatment effectiveness increases with increasing chemical dosage and pH. At a lime dosage of 400 mg/ℓ , fecal coliform, fecal streptococcus, and total coliform concentrations could be reduced by two orders of magnitude. It was also found that about 90 percent removal of bacteria can be achieved through alum clarification over a broad range of alum dosage.

Jar tests employing the f2 bacteriophage virus, lake water, and a variety of chemical coagulants and polyelectrolyte coagulant aids were conducted by York and Drewry (705). As shown in Table 38 , aluminum sulfate (alum), ferric chloride, ferric sulfate, ferrous sulfate, and polyelectrolyte B were found to give maximum virus removals greater than 90 percent at optimum dosage.

TABLE 38 . COMPARISON OF THE EFFECTIVENESS OF THE COAGULANTS TESTED (705)

Coagulants-Coagulant Aids	Dose mg/ℓ	Maximum Virus Removal percent
$Al_2(SO_4)_3$	25	99.9
$FeCl_3$	50	99.4
$Fe_2(SO_4)_3 \times H_2O$	50	92.0
$FeSO_4$ and $Ca(OH)_2$	36	93.5
$Al_2(SO_4)_3$ and $Na_2OAl_2O_3$	30 23	98.6
Polyelectrolyte A	2.0	76
$Al_2(SO_4)_3$ and polyelectrolyte B	18 1.0	99.2
Polyelectrolyte B	2.0	99.6
$Al_2(SO_4)_3$ and polyelectrolyte B	18 0.7	99.8
$Al_2(SO_4)_3$ and polyelectrolyte C	18 0.4	99.3
$Al_2(SO_4)_3$ and polyelectrolyte E	18 0.1	99.3
$Al_2(SO_4)_3$ and polyelectrolyte F	18 0.1	99.6
$Al_2(SO_4)_3$ and polyelectrolyte D	18 1.0	99.4

Berg et al. (56) experimentally mixed polio virus I and secondary effluent in containers, added lime, and stirred the solution for 15 min to allow formation of floc particles. Settling followed for 60 to 75 min. The removals obtained with varying dosages of lime are shown in Table 39 . Large coagulant doses were capable of effecting virus removals of up to 99.9 percent. After further investigation, the authors concluded that significant destruction of viruses can be attributed to the high pH occurring with high lime concentrations (in the range of 400 to 500 mg/ℓ).

TABLE 39. REMOVAL OF POLIO VIRUS 1 FROM SECONDARY EFFLUENT BY FLOCCULATION WITH $Ca(OH)_2$ (56)

$Ca(OH)_2$ Concentration mg/ℓ	Initial Virus Concentration pfu/ℓ	pH of Treated Effluent	Surviving Virus Concentration pfu/ℓ	Virus Removal percent
200	33,333	9.30	2,200	92.3
300	51,480	10.21	15,912	69.1
400	55,000	11.30	1,940	96.5
500	33,333	11.03	505	98.5
500	33,333	11.01	47	99.86

Chaudhuri and Engelbrecht (116) used water devoid of extraneous organic matter in their laboratory investigation of virus removal with alum. Using the experimentally determined optimum pH and dosage, 98.0 percent removal of bacteriophage T4 and 99.9 percent removal of bacteriophage MS2 were obtainable. However, the addition of organic matter in the form of albumin or treated wastewater lowered these efficiencies. For example, only 95.7 percent removal of bacteriophage T4 was obtained after the addition of 200 mℓ/ℓ of settled wastewater. Further experiments demonstrated no inactivation of the virus particles that were removed in the settled floc. However, Brunner and Sproul (86) demonstrated 60 percent permanent inactivation in the case of polio virus I removed from solution with aluminum phosphate precipitates. Their studies of polio virus I and bacteriophage T2 removal showed that under optimum conditions, removals can reach 98 and 94 percent, respectively, with aluminum and calcium precipitation. Actual removals are related to pH and chemical dosage.

Chemical treatment (high pH) holds considerable promise as a means of effectively inactivating or destroying pathogenic organisms contained in wastewater. By itself, chemical treatment cannot be relied upon to produce a pathogen-free effluent; used in conjunction with disinfection, however, it can help ensure that such an effluent is achieved.

TERTIARY TREATMENT: ION EXCHANGE

INTRODUCTION

The process of selective ion exchange has long been utilized in the treatment of industrial process waters and in domestic water supply softening. Ion exchange resins (539) are classified by the charge of the exchangeable ion. Thus, resins may be either catonic or anionic. General purpose resins will selectively exchange both cations and anions. The operational features of the ion exchange process are well developed and reliable. Such systems offer a reliable method of removing inorganic contaminants from the wastewater stream.

As can be seen by an examination of Table 40 , very little information is currently available on removals of contaminants from municipal wastewater by use of ion exchange techniques. The process has not been economically feasible for treatment of municipal wastewater. Several research programs focusing on the application of ion exchange to municipal wastewater treatment are presently under way. The most promising future application appears to be for ammonia or nitrate removals.

WATER QUALITY PARAMETERS

Eliassen and Tchobanoglous (194, 195) conducted a review of the literature. They found that removals of phosphorus and nitrogen by tertiary wastewater treatment incorporating ion exchange can reach 90 percent. The actual removal efficiency was seen to depend upon the type of preceding treatment. Evans (209) investigated the removal of nitrate by ion exchange, concluding that the strong acid/weak base ion exchange process is well suited for this purpose. With the exception of these few studies of phosphorus and nitrate, most research performed to date has focused on ammonium removal, since specific exchange resins are not available for either the phosphorus or nitrate ions. However, some zeolite exchange resins do have unusual selectivity for the ammonium ion. This fact has encouraged research activity.

On the basis of both pilot and laboratory scale investigations, it appears that effluent ammonia concentrations of less than 1 mg/ℓ can be expected with ion exchange (362, 438). In

TABLE 40. LITERATURE REVIEWED PERTAINING TO ION EXCHANGE

Contaminant	Reference Number
Water Quality Parameters	
Ammonia	3, 65, 90, 103, 150, 194, 233, 362, 438, 445, 517, 539, 694
BOD	64, 65, 103, 150, 255, 438
COD	64, 65, 103, 150, 196, 438, 707
Chlorides	196, 438
Nitrates	3, 65, 90, 194, 195, 209, 539, 694
Nitrites	90, 194, 694
Phosphates	64, 65, 103, 150, 194, 195, 196, 197, 209, 238, 438
Suspended solids	3, 64, 65, 103
Total dissolved solids	103, 121, 150, 233, 707
Total organic carbon	64, 65
Other	280, 318
Elemental Contaminants	
Arsenic	275, 506, 576
Boron	538
Cadmium	370, 391, 506
Chromium	370, 391, 506, 542, 549
Cobalt	370
Iron	370
Lead	370
Manganese	370
Mercury	370, 391, 452, 506

TABLE 40 (continued)

Contaminant	Reference Number
Elemental Contaminants	
Nickel	370
Selenium	391
Other	372, 506
Synthetic/Organic Contaminants	271, 541

a pilot plant study, Mercer et al. (445) used zeolite columns to test secondary effluent containing 10-19 mg/ℓ ammonia. Greate than 99 percent removal of ammonia was achieved. Similarly, 99. percent of the ammonia in activated carbon effluent was removed by a zeolite in laboratory scale experiments by McKendrick et al. (438).

The Environmental Protection Agency (517) reviewed pilot plant studies involving the use of clinoptilolite - a naturally occurring zeolite - for wastewater treatment. Ammonia removals ranged from 93 to 97 percent.

It should be noted that the ion exchange process using a zeolite such as clinoptilolite does not result in the production of a sludge containing the removed ammonia. Rather, the spent zeolite is regenerated with a lime slurry, which is subsequently air stripped, discharging ammonia to the atmosphere.

ELEMENTAL CONTAMINANTS

Ion exchange techniques have been principally applied for the removal of elemental contaminants from industrial waste streams (506). Few studies have dealt with the application of ion exchange techniques to municipal wastewaters for elemental contaminant removal. Lindstedt et al. (391) investigated trace metal removal and concluded that a cation-anion exchange sequence was effective in reducing the concentrations of cadmium, chromium, and selenium in secondary effluent. Removal efficiencies are summarized in Table 41.

TABLE 41 . TRACE METAL REMOVALS BY ION EXCHANGE (391)

Trace Metal	Percent Removal After Given Process Cation Exchange	Cation-Anion Exchange
Cadmium	99	99.9
Chromium	5	96
Selenium	1	99.7

BIOCIDAL CONTAMINANTS

Biocidal contaminant removal through ion exchange has also received little attention in the literature. In the only study located, Grover (271) stated that trifluralin, triallate, diallate, and nonionic herbicides were readily adsorbed on both cationic and anionic exchange resins, with somewhat more adsorption occurring on the cationic than on the anionic form.

NITROGEN REMOVAL PROCESSES

INTRODUCTION

Interim primary drinking water standards established by the EPA set a nitrate limit of 10 mg/ℓ in the nitrogen form. Nitrogen concentrations in raw municipal wastewaters generally exceed this value, ranging from 15 to 50 mg/ℓ. Unless facilities are specifically designed to remove nitrogen, much of it will remain essentially unaffected, passing through the varying stages of treatment to ultimately enter the environment. Moreover, reuse of wastewater treatment plant effluents for direct groundwater recharge, indirect groundwater recharge through land application, or indirect reuse as a potable water supply is on the increase. Such reuse policies make effective nitrogen removal an important aspect of any wastewater treatment scheme.

In raw municipal wastewater, nitrogen is primarily found in the form of both soluble and particulate organic nitrogen and as ammonium ions. Conventional primary and secondary treatment transforms some of this organic nitrogen into ammonium ions. Part of the ammonium ion is oxidized to nitrate, and about 15 to 30 percent of the total nitrogen is removed.

Tertiary treatment processes designed to remove wastewater constituents other than nitrogen often remove some nitrogen compounds as well. However, removal is often restricted to particulate forms, and overall efficiency is generally low. Two tertiary processes particularly designed to remove nitrogen have been developed: nitrification-denitrification and ammonia stripping. Tertiary nitrification-denitrification usually involves two stages. Nitrification occurs in an initial stage, during which ammonium ions are oxidized to nitrite and nitrate ions by nitrifying bacteria. These nitrite and nitrate ions are in turn reduced to nitrogen gas which simply escapes from the system.

Ammonia stripping is effective only in removing ammonia nitrogen from municipal wastewater and has no effect on organic nitrogen, nitrite, or nitrate. Several ammonia stripping plants are in operation in the U.S. (Lake Tahoe, California, Orange County, California), but the process has been found to be expensive. A number of technical problems remain to be solved as well (438).

Nitrification and denitrification are biological reactions which occur naturally during several conventional treatment processes such as activated sludge treatments, aerobic lagooning, and anaerobic digestion. The activated sludge process, in particular, can be closely controlled to promote nitrogen removal. Such treatment processes are not principally designed to remove nitrogen, and both nitrification and denitrification occur only as secondary reactions. For ease of reference, however, all literature reviewed on the general topic of nitrogen removal has been tabulated on Table 42, including tertiary processes specifically designed for this purpose.

WATER QUALITY PARAMETERS

There is general agreement that a system incorporating secondary biological treatment and tertiary nitrification-denitrification should achieve 80 to 95 percent total nitrogen removal at design flows (280, 483). The nitrification process alone removes only 5 to 10 percent of the total nitrogen entering the process, while oxidizing up to 98 percent of the ammonia nitrogen present to nitrate (236).

Average nitrogen data from systems incorporating nitrification-denitrification processes recorded by an EPA Technology Transfer Publication (236) is presented in Table 43 . Based on this report, the predicted effluent quality from a nitrogen-denitrification system will be 1.0 mg/ℓ organic nitrogen, 0.5 mg/ℓ ammonia nitrogen, 0.5 mg/ℓ nitrate nitrogen, and 2.0 mg/ℓ total nitrogen.

Nitrification is in itself an oxygen-demanding process and therefore reduces the total oxygen demand (TOD) in the wastewater effluent. Conventional biological or physico-chemical treatment obtaining up to 90 percent BOD reduction will only partially reduce the TOD of treated wastewater. For instance, such treatment will only reduce an influent TOD of 490 mg/ℓ to an effluent TOD of over 100 mg/ℓ. Nitrification will reduce the TOD of this effluent to less than 40 mg/ℓ (236).

Since the denitrification step involves the oxidation of carbonaceous material, a reduction in biochemical oxygen demand and total organic carbon can also be expected, in addition to the effective reduction of TOD.

Nitrogen removal by ammonia stripping was studied by O'Farrell et al. (492), who reported a 90 percent removal of ammonia from a non-nitrified, lime-clarified secondary effluent at pH 11.5. During a warm weather study performed at Lake Tahoe, stripping produced a 95 percent removal of ammonia nitrogen at pH 11.5 and using 400 cu ft of air per gallon of wastewater (280). Any arbitrary percentage removal can be achieved with this type of system within available engineering capabilities, although higher removals mean higher costs.

TABLE 42. LITERATURE REVIEWED PERTAINING TO NITROGEN REMOVAL

Contaminant	Reference Number
Water Quality Parameters	
Ammonia	3, 4, 6, 40, 66, 127, 194, 204, 236, 280, 328, 428, 438, 475, 483, 492, 517, 527, 539, 594, 620, 630, 684, 694, 706
BOD	40, 236, 330
Chlorides	403
Nitrates	3, 4, 40, 194, 195, 204, 236, 256, 257, 327, 328, 330, 428, 475, 483, 517, 527, 539, 594, 629, 630, 694
Phosphates	194
Suspended solids	40, 330, 629

TABLE 43. EFFLUENT NITROGEN CONCENTRATIONS IN TREATMENT SYSTEMS INCORPORATING NITRIFICATION - DENITRIFICATION (236)

	Average Effluent Nitrogen, mg/ℓ				
Type and Process Sequence	Organic-N	NH_4^+-N	NO_3^--N	NO_2^--N	Total N
Lime treatment of raw sewage, nitrification, denitrification	1.1	0.3	0.5	0.0	1.9
Primary treatment, high rate activated sludge, nitrification, denitrification, filtration	0.8	0.0	0.7	0.0	1.5
Primary treatment, roughing filters, nitrification, denitrification, filtration	0.8	0.9	0.6	0.0	2.3

SECTION 6

DISINFECTION: CHLORINATION

INTRODUCTION

Until recently, chlorination was considered virtually an unmixed blessing as a cheap, effective method to destroy bacteria and viruses. It is now recognized, however, that chlorination of wastewater may create chlorinated compounds harmful to the environment and to human health. The extent of this potential hazard has not yet been determined; new and existing wastewater treatment plants continue to utilize chlorine for disinfection. The primary purpose of municipal wastewater chlorination is the destruction of pathogenic microorganisms. This is reflected in the literature reviewed, shown in Table 44.

WATER QUALITY PARAMETERS

Zaloum and Murphy (711) concluded that chlorination of treated wastewater effluents does not reduce BOD, COD, and total organic carbon. Susag (1346), however, found BOD reductions by chlorination of up to 2 mg/ℓ per mg/ℓ of chlorine added. These values are somewhat misleading, in that BOD reduction was due both to oxidation of the organic material and to the formation of chlorinated organics resistant to bacterial action.

When chlorine is added to a wastewater containing ammonia nitrogen, ammonia reacts with the hypochlorous acid formed to produce chloramines. Further addition of chlorine converts the chloramines to nitrogen gas. The reaction is influenced by pH, temperature, contact time, and initial chlorine-to-ammonia ratio. If sufficient chlorine is added, 95 to 99 percent of the ammonia will be converted to nitrogen gas with no significant formation of nitrous oxide. The quantity of chlorine required was found to be 10 parts by weight of chlorine to 1 part of ammonia nitrogen when treating raw sewage. This ratio decreased to 9:1 for secondary effluents, and 8:1 for lime-clarified and filtered secondary effluent (627).

ELEMENTAL CONTAMINANTS

Little information is available on the minimal removal by chlorination of elemental contaminants. Andelman (16) studied

TABLE 44. LITERATURE REVIEWED PERTAINING TO CHLORINATION

Contaminant	Reference Number
Water Quality Parameters	
Ammonia	21, 36, 62, 65, 71, 360, 361, 517, 531, 597, 713
BOD	65, 171, 298, 499, 516, 569, 627, 711
COD	65, 516, 520, 627, 711
Chlorides	342
Cyanides	28, 499
Nitrates	65, 342, 517, 614
Nitrites	360, 361, 517
Phosphates	65, 342
Suspended solids	65, 516, 569
Total dissolved solids	569
Total organic carbon	65, 711
Others (general)	203, 205, 280, 360, 361, 437, 677
Elemental Contaminants	
Barium	16
Boron	638
Copper	16
Iron	360, 499
Manganese	360, 361, 559
Mercury	499, 692
Nickel	16, 361

TABLE 44 (continued)

Contaminant	Reference Number
Synthetic/Organic Contaminants	5, 42, 85, 102, 250, 334, 464, 536, 581, 596, 619, 669, 713
Biological Contaminants	
Adeno virus	49
Bacteria	6, 38, 88, 109, 110, 154, 158, 171, 208, 311, 332, 407, 604
Coliforms	88, 89, 158, 171, 195, 201, 208, 251, 307, 311, 360, 361, 454, 516, 604, 608, 647, 649, 673, 687, 689
Coxsacki virus (A&B)	49, 136, 198, 311, 439, 564
ECHO virus	198, 311, 585
Escherichia coli	75, 195, 201, 223, 311, 564, 656, 661
Fecal streptococci	158, 195, 307, 516
Hepatitis virus	311
Mycobacterium	223, 311
Parasitic worms	88, 311, 562, 604
Polio virus	88, 118, 198, 311, 401, 404, 564, 585, 601, 659
Protozoa	88, 201, 223, 621
Salmonella	88, 158, 208, 223, 514
Shigella	88
Vibrio cholerae	88
Virus	38, 48, 49, 50, 54, 55, 71, 88, 109, 110, 152, 153, 185, 200, 201, 208, 233, 259, 311, 332, 365, 368, 400, 401, 447, 463, 487, 514, 575, 584, 585, 604, 637, 664
Other (general)	88, 296

the effects of chlorination on barium, copper, and nickel. The treatment effected a 34-percent reduction in barium, a 5-percent reduction in nickel, and had no effect upon copper. Kokoropoulos (360) reported hypochlorous acid reacted with soluble iron (II) and manganese (II) to form precipitates.

SYNTHETIC/ORGANIC CONTAMINANTS

No research was found to address removal or destruction effects of chlorination on any of the synthetic/organic contaminants. However, considerable interest has recently developed concerning the production of chlorine-containing organic compounds by chlorination. The reactions of chlorine with organic compounds in water are diverse, including oxidation, substitution, addition, and free radical reactions. Chlorination may produce several different chlorinated products from a single organic pollutant molecule. Some of these compounds have been identified as toxic to aquatic life by Snoeyink (596), Brungs (85), and others.

Jolley (334) evaluated chlorine-containing organic constituents in chlorinated effluents and found that stable chlorine-containing compounds were present after effluents had been chlorinated to a 1 to 2 mg/ℓ chlorine residual. These compounds are identified in Table 45.

TABLE 45. IDENTIFICATION OF CHLORINE CONTAINING CONSTITUENTS IN CHLORINATED EFFLUENTS (334)

2 - Chlorobenzoic acid
3 - Chlorobenzoic acid
4 - Chlorobenzoic acid
8 - Chlorocaffeine
6 - Chloroguanine
3 - Chloro-4-hydroxybenzoic acid
4 - Chloromandelic acid
4 - Chloro-3-methylphenol
2 - Chlorophenol
4 - Chlorophenol
4 - Chlorophenylacetic acid
3 - Chlororesorcinol
5 - Chlorouracil
5 - Chlorouridine
8 - Chloroxanthine

A similar project was conducted by Glaze and Henderson (250). The chlorinated organics identified in this study are listed in Table 46.

Shimizu et al. (581) stated that halogenated nucleic acid bases are incorporated into the nucleic acid. Also, the incorporation of 5-deoxybromouridine in DNA and 5-fluorouracil into RNA are known to cause mutations. No work has been completed to determine how nucleic acids react with chlorine or the resulting mutations.

TABLE 46. CHLORINATED ORGANICS IN WASTEWATER EFFLUENT (250)

Chloroform	Dibromochloromethane
Dichlorobutane 3-chl	3-chloro-2-methylbut-1-ene
Chlorocyclohexane (-18)	Chloroalkyl acetate
O-dichlorobenzene	Tetrachloroacetone
P-dichlorobenzene	Chloroethylbenzene
Pentachloroacetone	Hexachloroacetone
Trichlorobenzine	Dichloroethyl benzene
Chlorocumene	
N-methyl-trichloroaniline	Dichlorotoluene
Trichlorophenol	
Chloro-a-methyl benzyl alcohol	
Dichloromethoxytoluene	
Trichloromethylstyrene	Trichloroethyl benzene
Dichloro-a-methyl benzyl alcohol	
Dichloro-bis (ethoxy) benzene	
Dichloro-a-methyl benzyl alcohol	Trichloro-N-methylanisole
Trichloro-a-methyl benzyl alcohol	Tetrachlorophenol
Trichloro-a-methyl benzyl alcohol	Trichlorocumeme
Tetrachloroethylstyrene	Trichlorodimethoxybenzene
Tetrachloromethoxytoluene	
Dichloroaniline derivative	
Dichloroaniline derivative	Dichloroacetate derivative
Trichlorophthalate derivative	
Tetrachlorophthalate derivative	

BIOLOGICAL CONTAMINANTS

The effectiveness of chlorination as a disinfection process has long been recognized. All researchers are in agreement that the effectiveness of disinfection by chlorine is influenced by time and chlorine concentration and also by: (1) whether the chlorine residual is free or combined; (2) effectiveness of mixing; (3) whether or not particulates are present; (4) pH; (5) temperature; and (6) the concentration, condition, and nature of the organisms. Keeping these limitations in mind, an idea of the relative resistances of organisms to disinfection by chlorine can be seen in Table 47.

TABLE 47. EFFECT OF CHLORINATION ON VARIOUS ORGANISMS (311)

Group	Organism	Chlorine Residual (mg/ℓ)	Time min.	Efficiency
Virus	Infectious Hepatitus	1	30	Survived
		15	30	Inactivated
	Coxsackie	5	2.5	Survived
	Coxsackie	1.0	3	99.6% Inactivated
	Echo	1.95	6.5	Survived
	Poliovirus I	0.53	14	Survived
	Coliphage B	0.03	10	20% Survival
	Theiler Phage	0.03	10	Inactivated
Bacteria	*M. tuberculosis*	1-5	120	99% Kill
		2	30	99% Kill
		1	30	Destroyed
	E. coli	0.14	3	99.9% Kill
	Coliforms	0.03	10	52% Kill
		1-1.2	15	99% Kill
	Total Count	trace	15	98-99% Kill
Nematodes	Diplogaster	2.5-3	120	Survived and
	Cheilobus	15-45	1	Mobile
Others	*S. mansoni* (ova and miracidia)	0.2-0.6	30	Killed
	S. japonicum (ova and miracidia)	0.2-0.6	30	Killed

Eliassen and Tchobanoglous (195) found that 2 to 6 mg/ℓ of chlorine applied for 20 min would effect a 99.99 percent kill of the total coliforms, fecal coliforms, and fecal streptococci present in wastewater influent.

The effect of chlorination on entamoebic cysts was investigated by Stringer and Kruse (621). It was concluded that the hypochlorous acid (HOCl) form of free available chlorine was the most rapidly acting cysticide compared with other halogen species. Entamoeba histolytica cysts and tapeworm eggs withstand the chlorination treatment generally applied to waste treatment effluent (88). Rudolfs et al. (562) reported the development of active embryos from a majority of ascaris eggs in contact with chlorine solutions for 30 min.

Davis and Keen (158) conducted a research project on the ability of municipal wastewater bacteria to survive chlorination and reestablish populations after discharge. Fecal coliform, fecal streptococci, and total coliform enumeration was performed with further differentiation into lactose nonfermenters within and outside the family Enterobacteriaceae. It was determined that the vast majority of the wastewater bacterial species that reestablished their populations within 21 days following chlorination were lactose nonfermenters not included in the enterobacteriaceae. Many of these bacteria could be pathogenic under the appropriate conditions and may constitute a threat to public health in receiving waters designated for contact recreation.

Virus inactivation is one of the more difficult tasks of chlorination, but as in any disinfection process, required kills can be achieved by lengthening the time or increasing the concentration (311). A study of the inactivation of viruses in wastewaters by chlorination was performed by Lothrup and Sproul (401). It was ascertained that:

- High-level inactivation of viruses can be obtained in treated and untreated domestic wastewaters. Present chlorination practices (1 mg/ℓ of residual), however, are inadequate for a high level of virus inactivation.
- A combined chlorine residual of 28 mg/ℓ was required to produce a 99.99 percent inactivation of the T2 bacteriophage in settled raw wastewater after a 30-min contact time.
- A combined chlorine residual of 40 mg/ℓ was required to provide a 99.99 percent destruction of the Type 1 polio virus in settled wastewater after a 30-min contact time.
- Free chlorine residuals of 0.2 to 0.4 mg/ℓ , after 30 min, produced a complete inactivation of the polio virus and T2 phage in the secondary effluent.

- In experimental runs with synthesized storm-water overflow samples, a 100 percent inactivation of the Type 1 polio virus was obtained by providing a free chlorine residual.

- The T2 bacteriophage was much less sensitive to combined chlorine residuals than are the coliform organisms and somewhat more sensitive than the polio virus to combined chlorine residuals.

A similar research project investigating the inactivation of enteroviruses by chlorination was conducted by Shuval et al. (585). The following conclusions were obtained:

- The strain of ECHO virus used was sensitive to the combined chlorine in the sewage, with reductions of 99 percent in 30 min and 99.93 percent in 6 hr using 3.6 mg/ℓ of chlorine. No virus was recovered in the sample tested after 4 hr contact with 7 mg/ℓ of chlorine dosage or after 2.5 hr with 11 mg/ℓ of applied chlorine. Inactivation was shown to be a function of time and chlorine concentration under the test conditions.

- The strain of polio virus Type 1 used was much less sensitive to the combined chlorine, showing only 50 and 90 percent reductions in 6 hr using 4 and 11 mg/ℓ chlorine dosage respectively.

- Coliform reductions obtained followed known patterns, with a standard of less than 100 coliforms/100 mℓ being obtained in 80 percent of the samples after 2 hr of contact with a chlorine dose of about 8 mg/ℓ.

The minimum concentration of chlorine required for complete inactivation of the Sabin oral poliovaccine Type I virus strain was examined by Varma et al. (659). Various exposure periods with pH 5.2 at 20°F were studied. A concentration of 22 mg/ℓ for 5 min of exposure time, 19 mg/ℓ for 15 min, 19 mg/ℓ for 30 min, 17 mg/ℓ for 45 min, and 14 mg/ℓ for 60 min. Nonetheless, on the basis of the literature surveyed, it is evident that chlorination per se does not provide conclusive proof of disinfection.

Boardman and Sproul (71) described the protection afforded viruses associated in particulate matter. Surface adsorption gave no viral protection. When viruses are embedded within particles, the disinfecting molecules must diffuse through the particle matrix before reaching the virus and initiating any inactivation. Chemical diffusion is a slow process; as a consequence, virtually all the embedded viruses are protected from disinfection.

DISINFECTION: OZONATION

INTRODUCTION

Ozone, an allotropic form of oxygen, is a powerful oxidizing agent for the disinfection of wastewater. Ozone is used in over 100 municipalities in Europe for disinfection of drinking water. Certain chemical features make ozone treatment a particularly attractive method of water purification:

- It is a powerful oxidant which reacts rapidly with most organic compounds and microorganisms in wastewater.
- It does not impart taste and odor to potable water.
- It is produced from oxygen in air by means of electric energy.

On the negative side, the cost of ozonation is not presently competitive with chlorine disinfection. Moreover, long-term residual disinfection capabilities are lacking, and the instability of ozone generally necessitates its generation on site (660).

The principal ozone decomposition products in aqueous solution are molecular oxygen and the highly reactive free radicals HO_2^-, OH^-, and H^+. Very little is known about the significance of the free radical intermediates on the germicidal properties of ozone solutions. The same free radicals are produced by irradiation of water, and it has been reported that HO_2^- and OH^- radicals contribute significantly to the killing of bacteria by this process.

As seen in Table 48, a considerable amount of information is available on the destruction of various pathogens by ozonation, however little information was found on the effect of ozone upon other contaminants.

WATER QUALITY PARAMETERS

Because of its strong oxidizing character, ozone is very reactive toward the organic compounds which make up the BOD, COD, and the total organic carbon. Under ideal conditions the reactions would result in almost complete oxidation and only carbon dioxide as a reaction product. In practice, ozonation results

TABLE 48. LITERATURE REVIEWED PERTAINING TO OZONATION

Contaminant	Reference Number
Water Quality Parameters	
Ammonia	6, 244, 332, 353, 545
BOD	170, 171, 353, 427, 463, 474
COD	127, 170, 244, 351, 474, 571
Nitrates	244, 332, 474, 545
Nitrites	244, 332, 353, 545
Phosphates	474
Suspended solids	244
Total organic carbon	474, 587
Others (general)	207
Elemental Contaminants	227, 228
Biological Contaminants	
Adeno virus	49, 110
Bacteria	6, 34, 110, 170, 171, 243, 283, 351, 353, 556
Clostridium botulinium	660
Coliforms	171, 244, 351, 402, 673
Coxsackie virus (A&B)	110
ECHO virus	110
Escherichia coli	34, 49, 343, 344, 351, 660
Fecal streptococci	34, 351
Parasitic worms	444, 660

TABLE 48 (continued)

Contaminant	Reference Number
Biological Contaminants	
Polio virus	110, 118, 343, 344, 409
Salmonella	351
Virus	49, 50, 51, 110, 152, 170, 259, 400, 402, 508, 509
Other (general)	343

in only partial oxidation and produces simpler organic molecules. Both Ghan and Nebel reported COD removals of less than 40 percent. Morris found that the apparent BOD of a wastewater can increase after ozonation as a result of refractory organic molecules being oxidized to simpler, biodegradable compounds. If the ozone is applied after other treatment processes (as is normal), the increase in organic nutrient molecules can lead to the growth in the distribution system of algae, slime bacteria, and the possible regrowth of any pathogens not destroyed during treatment.

Ozone is effective at decreasing concentrations of organic suspended solids and organic nitrogen through oxidation. Ozonation can assist in suspended solids removal through froth flotation mechanisms induced through the process. Ozone will also oxidize nitrites to nitrates, but will not react with ammonia (332). There is little evidence to date that ozonation will produce any toxic or carcinogenic oxidation by-products as will chlorination.

ELEMENTAL CONTAMINANTS

Furgason and Day (228) studied the feasibility of ozonation for iron and manganese removal from raw water with relatively high input concentrations. The study demonstrated that ozone effectively oxidized the iron and manganese to an insoluble form which could be filtered from the water.

Complete oxidation of the minerals required a reaction time of 30 sec. Filtration studies indicated a relatively fine medium was required to remove the oxide precipitate.

BIOLOGICAL CONTAMINANTS

The use of ozone as a wastewater disinfectant was reviewed by Venosa (660). It was concluded that with 0.1 mg/ℓ of active chlorine, 4 hr would be required to kill 6×10^4 E. coli cells in water, whereas with 0.1 mg/ℓ of ozone only 5 sec would be necessary. When the temperature was raised from 22°C to 37°C, the ozone inactivation time decreased from 5 sec to 0.5 sec. These investigations revealed that the contact time with ozone necessary for 99 percent destruction of E. coli was only one-seventh that observed with the same concentration of hypochlorous acid. The death rate for spores of Bacillus species was about 300 times greater with ozone than with chlorine.

In the same study, Venosa also described bacteriological studies performed on secondary effluent from an extended aeration pilot plant in the Metropolitan Sewer District of Louisville, Kentucky. Using an average applied ozone dosage of 15.2 mg/ℓ for an average contact time of 22 min, fecal coliform reductions of greater than 99 percent were achieved, resulting in a mean fecal coliform concentration of 103 cells/100 mℓ , a mean total coliform concentration of 500 cells/100 mℓ , and a mean fecal streptococci concentration of 8 cells/100 mℓ in the final effluent. Laboratory results with raw sewage indicated that ozone could be successfully used to sterilize sewage containing *Bacillus anthracis*, influenza virus, and *B. subtilis* morph. *globibii*, and to inactivate toxins of *Clostridium botulinum*. Ozone consumption was 100 to 200 mg/ℓ for 30 min. Finally, Venosa found ozone to be many times more effective than chlorine in inactivating poliomyelitis virus. Identical dilutions of the same strain and pool of virus, when exposed to 0.5 to 1.0 mg/ℓ of chlorine and 0.05 to 0.45 mg/ℓ of ozone, were devitalized within 1.5 to 2 hr by chlorine, while only 2 min of exposure were required with ozone.

Majumdar et al. (409) also studied the inactivation of polio virus by ozonation and concluded that the inactivation is not totally complete. Results are summarized in Table 49 .

TABLE 49 . SURVIVAL OF POLIO VIRUS IN OZONATION CONTINUOUS FLOW STUDIES (409)

Type of Wastewater	Ozone Concentration (mg/ℓ)	Residence Time (min)	Average Survival (percent)
Primary wastewater	0.84	8.0	1.820
	1.47	2.0	0.016
	4.44	1.0	0.006
Secondary wastewater	0.79	8.0	2.055
	1.77	2.0	0.013
	5.05	1.0	0.006

Pavoni and Tittlebaum (508) recently studied ozone disinfection of viruses in the Fort Southworth Pilot Plant of the Metropolitan Sewer District in Louisville. Using F2 bacteriophage as the model virus, they demonstrated virtually 100 percent inactivation efficiency in the secondary effluent after a contact time of 5 min at a ozone dosage of approximately 15 mg/ℓ and a residual of 0.015 mg/ℓ . Of particular interest was the observation that the rate of inactivation was greater for F2 bacteriophages than bacteria. In addition, the following conclusions were reached:

1. F2 virus concentrations were shown to be unaffected by the flow or mixing of the ozone reactor.

2. F2 virus was inactivated with virtually 100 percent efficiency after a contact time of 5 min at a total ozone dosage of approximately 15 mg/ℓ and a residual of 0.015 mg/ℓ .

3. *E. coli* bacteria and F2 virus were inactivated with virtually 100 percent efficiency after a contact time of less than 15 sec in the absence of ozone-demanding material.

4. An extremely small number of viral particles was observed in effluent studies.

5. Oxidation by ozone appears to be the mechanism of kill for bacterial cells and viral particles. Ozone is theorized to act as a general oxidant causing cell lysis and the release of soluble COD.

Mercado-Burgos et al. (444) examined the effect of ozone on Schistosoma ova, concluding that the process was ineffective.

SECTION 7

LAND/GROUNDWATER

INTRODUCTION

Hundreds of municipal and industrial wastewater treatment plants dispose of their effluents to the land, as illustrated in Figure 2 . Thousands of wastewater lagoons percolate effluents into the ground. Millions of septic tank systems leach their wastewater into the ground. Most of these wastewaters travel through the soil and eventually reach groundwater aquifers. Unplanned groundwater recharge with wastewaters, therefore, must be recognized as occurring on a large scale.

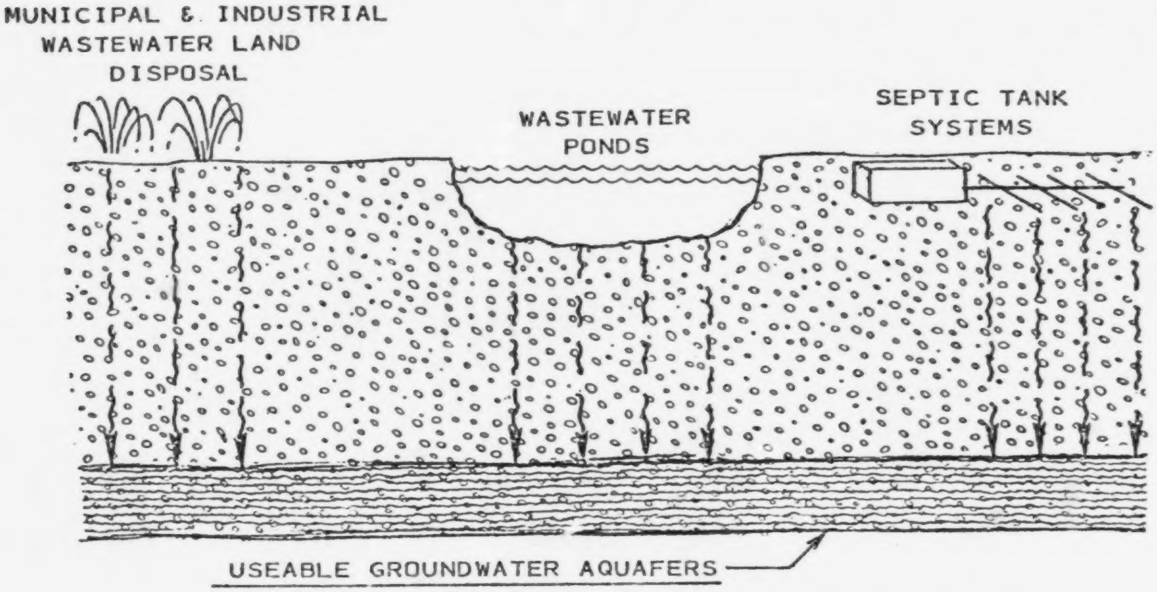

MUNICIPAL & INDUSTRIAL WASTEWATER LAND DISPOSAL
WASTEWATER PONDS
SEPTIC TANK SYSTEMS
USEABLE GROUNDWATER AQUAFERS

Figure 2. Unplanned wastewater reuse exists for many groundwater supplies.

The present interest in planned regulated wastewater reuse projects is bringing into focus an informal practice which has existed "in the closet" for a long time. The question is not whether wastewater reuse is acceptable, but rather how best to control what is an existing practice.

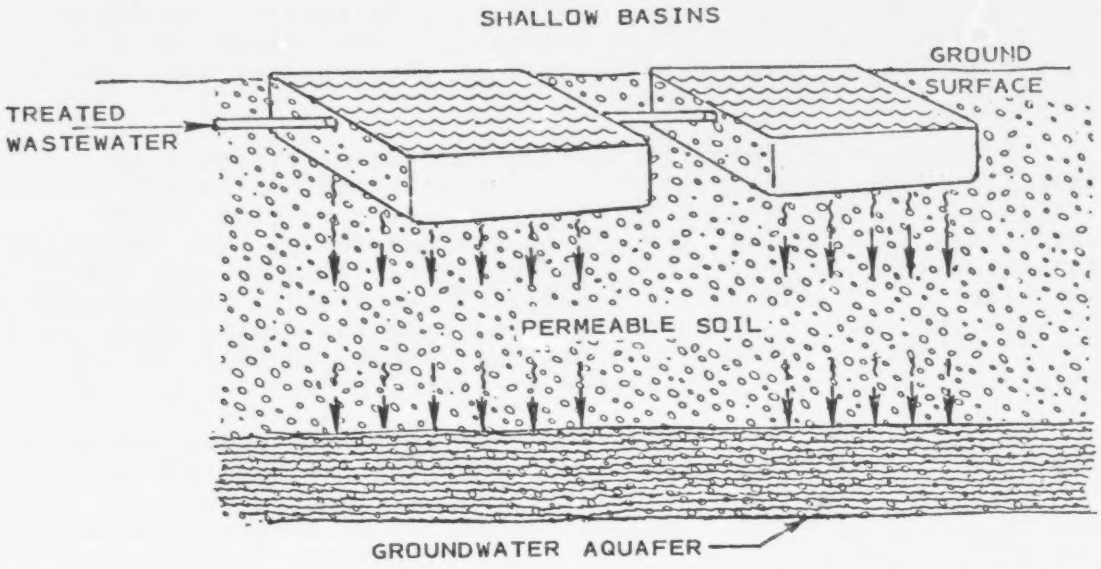
Groundwater recharge with treated municipal wastewater is accomplished by either planned or unplanned processes. Planned processes consist of two basic methods (Figure 3). The simplest and most widely used consists of conveying the treated effluent to shallow spreading basins and allowing the water to percolate through the soil to the groundwater. The second planned method consists of conveying the effluent to a well field and injecting the water directly into the aquifer. The major intent of these formal processes is to replenish groundwater basins, to establish saltwater intrusion barriers in threatened coastal aquifers, or to provide further treatment for ultimate extraction and reuse.

Unplanned recharge, as previously noted, accounts for much greater amounts of wastewater reaching groundwaters and includes percolation from irrigation and holding ponds and leachate from septic tanks.

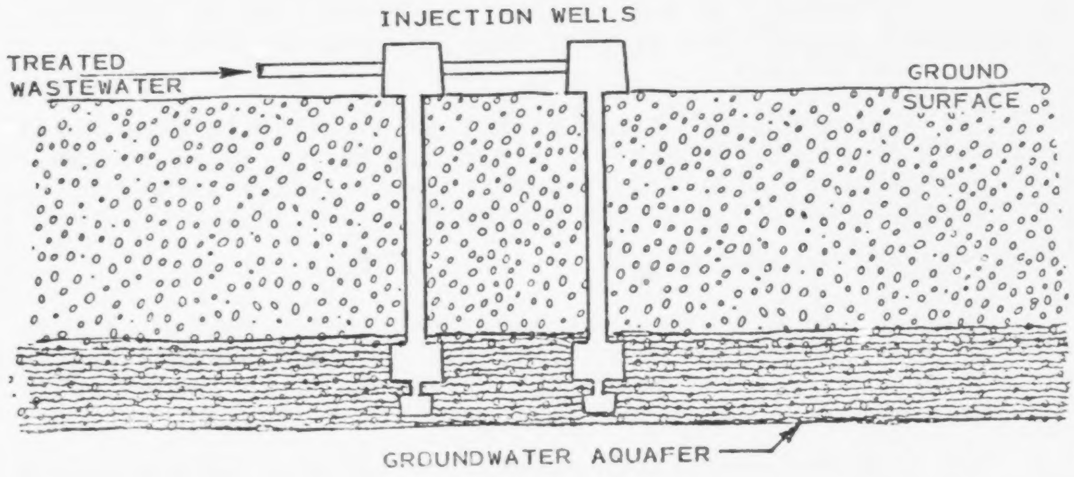
Many variables affect the potential for a public health hazard from land disposal. These include:

- The characteristics of the wastewater disposed;
- The rate of waste application;
- The hydrogeological characteristics of the disposal site;
- The method of disposal, e.g., crop irrigation, land spreading, percolation ponds, sanitary landfill, etc.;
- Proximity of public access;
- Utilization of groundwater potentially affected; and
- Local climate.

The above listed variables are not mutually exclusive; they interact in many ways to influence the rate and extent of transport of contaminants from each source and to influence the importance of each potential pathway. It is beyond the scope of this report to delve deeply into the science of waste application technology. This subject is being researched


SHALLOW BASINS
GROUND SURFACE
TREATED WASTEWATER
PERMEABLE SOIL
GROUNDWATER AQUAFER

PLANNED RECHARGE BY PERCOLATION


INJECTION WELLS
TREATED WASTEWATER
GROUND SURFACE
GROUNDWATER AQUAFER

PLANNED RECHARGE BY DIRECT INJECTION

Figure 3. Methods of planned recharge.

heavily by the EPA and other agencies to establish guidelines for the safe land disposal of effluents. This report will, however, discuss the current knowledge about associated potential public health problems.

Literature reviewed concerning wastewater disposal to land has centered on groundwater elemental and biological contaminants, and water quality parameters providing plant nutrients, such as nitrogen and phosphorus forms. The increased use of wastewater application to land has created a need for increased research into the dynamics of wastewater contaminants in soil systems. It is necessary to study the capabilities of these systems to destroy, neutralize, remove, concentrate, or otherwise affect applied wastewater contaminants.

A number of factors determines the degree to which groundwater may be contaminated by wastewater that is applied to land. Depth to the groundwater table and distance to an extraction point affect residual levels of phosphorus, bacteria, and other constituents for which removal appears to be a function of travel distance. Soil characteristics, native groundwater quality, assimilation capacity of the aquifer, and method of waste application also determine groundwater degradation and consequent health problems (568). Cation exchange and adsorptive capacities important in the removal of metal ions and viruses and of trace organics and solids are determined by soil composition. Porosity regulates infiltration rates to some extent, affecting contaminant residence time in surface layers. Residence time may, in turn, determine aerobic or anaerobic conditions.

Total groundwater volume cannot necessarily be considered an effective diluting agent. Uniform diffusion of recharged water cannot be guaranteed, and water quality may vary considerably both in area and in depth.

WATER QUALITY PARAMETERS

Research in this area has been principally concerned with nitrogen and phosphorus forms entering groundwater as a result of land application of wastewater. Removal of suspended solids from wastewater effluent has also received attention.

The problems and transformations associated with nitrogen forms in soils are relatively well known. Organic nitrogen and ammonia, when applied to soils under normal aerobic conditions, are rapidly converted by nitrifying bacteria to nitrite and nitrate. Both of these anionic forms move through soils in percolating water with little difficulty. Under anaerobic soil conditions, on the other hand, the process of conversion to nitrite and nitrate is inhibited. Ammonium ions and free ammonia persist and are held near the soil surface by adsorption onto soil particles, by cation exchange reactions, or by fixation in clay lattices.

In an acidic environment, nitrite has been found to react with secondary amines to produce nitrosamines. These compounds have recently been labeled carcinogenic, teratogenic, and mutagenic. The health hazards associated with nitrite and other forms of nitrogen in drinking water and crops have been delineated by the U.S. Department of Agriculture (663).

The most definitive study of nitrogen removal by land application of wastewater effluent was conducted by Herman Bouwer at Flushing Meadows, Arizona (73). He found that short flooding periods (two days flooding followed by five days drying) did not provide sufficient time to develop the anaerobic conditions for nitrate denitrification. A longer flooding period of ten days followed by two weeks of drying proved to be more favorable. With this schedule, oxygen in the soil was depleted during flooding, causing nitrogen (in the ammonium form) to be adsorbed by the clay and organic particles. Flooding was stopped before the cation exchange complex in the soil was saturated with ammonium. Upon drying, oxygen entered the soil, and ammonium was nitrified under aerobic conditions to nitrate. Concurrently, some of the nitrate formed was denitrified, in micro-anaerobic pockets in the otherwise aerobic upper soil zone, to nitrogen gas that escaped to the atmosphere. When flooding was resumed, if the basins were immediately flooded to a depth above 1 ft, the nitrates were quickly leached out of the top few feet of the soil to groundwater. However, if initial flooding was shallow (a few inches deep), the lower head allowed a low infiltration rate, a larger nitrate retention time in the microbiologically active soil zone, and further denitrification. At these lower initial hydraulic loading rates, nitrogen removals were as high as 80 percent. However, if high application rates were consistently maintained, nitrogen removal was only 30 percent with a peak nitrate surge to the groundwater after the start of each new flooding cycle.

A study by Preul (532) in 1966 provided the following observations of the movement and conversion of nitrogen in soil and the potential dangers of nitrate contamination:

1. Biological oxidation is the dominant mechanism affecting ammonia nitrogen as it passes through the soil. This action initially occurs at a high rate, and to a large extent, within several feet of the point of release of the septic tank effluent, if soil conditions are well aerated.

2. Nitrate contamination of groundwaters is a serious threat from shallow soil adsorption systems. High concentrations of ammonia nitrogen in septic tank effluents are quickly nitrified to high concentrations of nitrate, which pollute the groundwater. Dilution from groundwater or soil moisture and possibly denitrification aid in the deterrence of nitrate.

3. The effectiveness of adsorption in deterring the travel of nitrogen is limited, because of the rapid conversion of ammonia to nitrate. Laboratory experiments have shown that ammonium can be readily removed in soil by adsorption; but, under aerated soil circumstances, nitrification of these ions occurs before the flow can contact a sufficiently effective volume of soil.

Similarly, results of a study by Chapman et al. (115) have shown that, in Texas, irrigation with a sewage effluent was a potential source of nitrate pollution of the local groundwater. The results indicated that nitrification of ammonia nitrogen in the effluent is rapid and complete, taking place within the top 3 ft of soil. It was concluded that substantial amounts of nitrate would not be fixed by the soil and that, at a 3-in per week application rate, appreciable amounts of high nitrate water would percolate to the groundwater. Selective crop production of grains and grasses having high nitrogen uptakes (corn, bermuda grass, oats) was deemed the most effective method of protecting the groundwater.

Short daily flooding schedules evidently result in highly efficient nitrification to nitrate. Mc Michael and McKee (441) reported data from test basins in the Whittier Narrows and the Rio Hondo spreading grounds near Los Angeles, California. These basins, equipped to collect water at 2-, 4-, 6-, and 8-ft depths, received treated wastewaters on a daily basis using a short period of flooding and a longer period of drying. This cycle ensured completely aerobic conditions. Studies indicated that almost all the nitrogen had been converted to the nitrate form at depths of 8 ft.

Intermittent and continuous spreading of secondary effluent at the Hyperion Treatment Plant (572), in Southern California, resulted in the nitrogen transformations shown in Table 50.

TABLE 50. NITROGEN TRANSFORMATIONS RESULTING FROM DIFFERENT SPREADING TECHNIQUES (572)

	Organic-N	Ammonia-N	Nitrate-N
I. Under continuous spreading:			
Effluent applied (mg/ℓ)	7.1-8.5	15.5-17.5	0.2- 0.8
Percolate from 7-10 ft below ground (mg/ℓ)	1.2-2.7	8.7-18.5	5.2-18.1
II. Under intermittent spreading:	Organic-N	Ammonia-N	Nitrate-N
Effluent applied (mg/ℓ)	2.3-3.6	4.9-27.9	0.1-14.2
Percolate from 7-10 ft below ground (mg/ℓ)	1.1-2.1	0.0- 0.7	8.4-22.4

A study (572) conducted at the sewage farm in Arroyo Grande, California, indicated the pulse-like effect of intermittent spreading on the nitrate level in the percolate. During weekly flooding of the field with settled sewage effluent, the upper foot of the soil adsorbed 2,840 lb of organic and ammonia nitrogen/ac. As the fields drained and were exposed to oxygen, this adsorbed nitrogen was rapidly converted to nitrate, resulting in nitrate levels of 1,000 to 2,000 mg/ℓ in the soil solution of the top foot. Consequently, the subsurface drainage water exhibited pulses of high nitrate concentration when flooding began. After all the nitrate was leached from the topsoil by the applied wastewater, the nitrate content of the drainage water returned to a low level.

When the South Tahoe Public Utilities Department sprayed treated sewage on forested hillsides in the fall of 1963, nitrogen removals by the soil mantle were more than 65 percent. The removals dropped to 26 percent in the winter when the ground was frozen. Significant amounts of ammonium ion were present in the upper 4 in of soil, but nitrate levels were low at all depths. The removal of nitrogen was attributed to the denitrification in the soil mantle under anaerobic conditions (572).

The California State Health Department (572) reviewed nitrogen removals and management at a number of well-injection waste disposal/recharge systems. Results of a six-month study on the injection of slow sand filter effluent are shown in Table 51.

TABLE 51. NITROGEN TRANSFORMATION IN RECHARGE AQUIFER, MG/ℓ (572)

	Organic-N	Ammonia-N	Nitrite-N	Nitrate-N
Natural water in the aquifer	.4-.5	.2	0.0	.4-4.8
Filter effluent before injection	2.2	1.5	.01	21.3
Recharged water in aquifer, 20 ft from injection well	1.4	1.2	.21	18.2
Recharged water mixture in aquifer, 500 ft from injection well	.9	.8	.003	6.1

As can be seen from this table, intermittent spreading techniques maintained aerobic conditions in the soil, making possible the oxidation of ammonia and organic nitrogen to nitrate. Under continuous spreading, anaerobic conditions prevailed, and ammonia was still present in significant concentrations at a 7- to 10-ft depth.

When a low-rate application system is used, the amount of nitrogen applied to soil with sewage effluent is not much more than can be removed by crops, according to Bouwer (72). A low-rate system involving application to wheat at 2.5 and 5 cm/week was cited. At the lower application rate, 92 percent of the nitrogen in the wastewater was removed in the soil, while at the 5 cm/week rate, 60 percent was removed. On the other hand, when animal waste slurries or other wastewater with a relatively high nitrogen content are applied, the amount of nitrogen supplied may far exceed that which can be utilized by crops. To obtain significant nitrogen removal under these circumstances, Bouwer suggests that the system be designed to stimulate denitrification in the soil. He cites an instance where this was done by installing an artificial barrier to water movement at a depth of 2 m, causing the formation of an anaerobic region. Ammonia and organic nitrogen in applied wastewater were converted to nitrate in the upper, aerobic region of the soil, which was then denitrified in the lower, anaerobic zone. The system removed 96 to 99 percent of the total nitrogen applied at rates of 1 to 2 cm/day.

Analysis of data in Table 50 indicates that the decay of nitrate in the aquifer was due to some extent to denitrification, as well as dilution. Considering such decay, the nitrate nitrogen level was expected to fall to 10 mg/ℓ before traveling a distance of 500 ft in the aquifer. Further tests revealed an anaerobic, microbiologically active zone in the aquifer in the vicinity of the injection well. During the tests, nitrate was largely removed by microbial denitrification within 150 ft of the injection well.

In summary, data indicate that, under proper management conditions, land application of wastewater effluent offers the potential to efficiently remove nitrogen from wastewater and protect groundwater from nitrate contamination. The most successful programs stressed an appropriate flooding/drying schedule to promote both aerobic nitrification and anaerobic denitrification processes, in order to ultimately convert ammonia nitrogen in the wastewater to nitrogen gas. However, if not properly managed, a definite danger exists of polluting groundwater resources with excess nitrates.

Recent studies of land application of wastewater effluent indicate that the soil system is highly efficient in removing phosphates from wastewater. Phosphate removal is both a function of soil composition and travel distance. In most soils, phosphorus not taken up by plants is immobilized due to the adsorption of phosphate onto the soil. Adsorption is followed by precipitation into various forms of calcium phosphate if the soil is basic (72). These reaction products are sufficiently insoluble, so that phosphorus is held in the upper few centimeters of most soils, and very little phosphorus moves into the groundwater (388). However, in the case of acidic, sandy soils with no iron or aluminum oxides, little phosphate is fixed. Thus, it may be necessary to remove phosphorus from wastewater before its application to such soils (72).

Hook, Kardos, and Sopper (299) reported that, under proper management, most of the phosphorus in wastewater remains in the soil at the disposal site or leaves as a nutrient in harvested crops. They found that soils differed in their abilities to retain phosphorus. In a heavy-textured soil high in iron and aluminum oxides and hydroxides (sesquioxides), phosphorus from effluent irrigation did not increase in the soil below a depth of 1 ft after 7 yr of irrigation. In a light-textured soil with half as much sesquioxides, phosphorus content increased to a depth of 3 ft after 6 yr of treatment.

Phosphorus removals at Flushing Meadows, Arizona (73), were found to be basically dependent upon the distance traveled by the wastewater through the soil. The chief removal mechanism was precipitation as calcium phosphate or magnesium ammonium phosphate, since the soils tested contained little iron, aluminum oxides, or other phosphate fixing materials. Underground travel distances of 30 ft produced 50 percent reduction; distances of several hundred feet were found to be sufficient for 90 percent phosphorus removal. However, the capacity of the soil to remove phosphorus decreased after the start of the project, holding stable at approximately 50 percent removal.

The fact that soils gradually lose their capacity to adsorb phosphates over long-term application is substantiated by Barrow's detailed analysis of this phenomenon (30). He concluded that previously applied phosphate had been converted to a form that was occupying phosphate adsorption sites, thus reducing the capacity of the soil to further adsorb phosphate.

Continuing studies at Lake George, New York (22), have shown that significant amounts of nitrates appear to reach the waters tributary to the lake. However, the wastewater land treatment system appears to remove essentially all phosphorus, thus reducing the potential for algal bloom in the lake.

Dugan et al.(185), in their work on land disposal of wastewater in Hawaii, noted that phosphorus removals of over 95 percent within a 5-ft depth of percolation were obtained when secondary wastewater was applied to grassed areas.

Suspended solids are removed very effectively by land application systems. In fact, one problem encountered in the spreading of wastewater is that nearly all suspended solids are filtered out in the top few inches of soil. This can cause clogging of soil pores and reduction of infiltration rates. There is no danger of groundwater contamination in applied wastewater from suspended solids. Literature data on suspended solids are not extensive, because experience has shown land application to be capable of removing virtually all solids at the surface. A study at Whittier Narrows, California (572), showed suspended solids removal, due to percolation, of 95 percent. At Flushing Meadows (73), the suspended solids concentration of the percolate was essentially zero, even though the solids in the wastewater applied reached 100 mg/ℓ. Similar findings were reported at Lake George (22), where virtually all BOD and suspended solids were removed from percolated effluent.

Results from a spray irrigation and runoff system used to dispose of a cannery waste (45) showed that even with a runoff-type system, suspended solids removals averaged 97 percent. Dugan et al. (185) reported similar high suspended solids removals with application of secondary effluent to Bermuda grass in Hawaii.

ELEMENTAL CONTAMINANTS

Municipal wastewater contains small amounts of nearly all metals. The degree to which a particular soil will protect underlying groundwater through removal of contaminants is primarily determined by the chemical and physical composition of the soil. Removal can occur through such processes as precipitation of solid phases, ion exchange, and adsorption. These processes are in turn controlled by soil pH, the oxidation/reduction potential, clay content, the presence and type of organic material, and the extent of soil saturation.

The general nature of reactions of sewage wastes with soil is well known. With time, wastes applied to land are broken down, and the dissolved constituents become part of the soil solution. Released cations can exchange with those already on exchange sites in the soil. Metals as ions or in the colloidal state can be adsorbed onto soil surfaces. When the levels of ions in solution exceed the solubility of corresponding solid phase compounds and minerals, those compounds can precipitate. When the solubility of solid phase compounds and minerals exceeds the levels of corresponding ions in solution, the compounds can dissolve. Constituents are also ingested by soil microorganisms and incorporated into soil organic matter.

Ions that are not removed by any of these processes, but that remain in the soil solution, are available for uptake by plant roots or leaching by water moving through the soil profile. Lindsay (388) studies the composition of the soil solution, concluding that it is controlled by the solubilities of solid phases. Thus, precipitation and dissolution reactions determine the activity of ions in solution, which in turn governs ion exchange.

Lindsay (388) also recognized the importance of the formation of metal-organic complexes and chelates in increasing the solubility and mobility of metals in soils. Brown (79) stressed this point. He found that it is misleading to predict the availability of metals in soils from their solubility in distilled water. He cites evidence that plant uptake of metal ions could not be predicted by the water solubility of the solid compounds. This may be because the soil solution in the vicinity of roots, unlike distilled water, is mildly acidic and contains organic metal-complexing agents (388).

All of the trace elements for which water quality criteria have been established may occur as either soluble or insoluble metal-organic complexes (615). Low molecular weight organic molecules tend to increase the penetration of complexed metal ions into the soil, while high molecular weight molecules and their complexed ions may be filtered out by the soil (72).

The chemistry of the metal organic complexes is complicated, and present knowledge of organic forms of the elements is insufficient to generalize. Where concentrations of trace elements in soil solutions are in excess of those predicted from inorganic solubility product considerations, the element is thought to occur in organic form. Because of this lack of knowledge, the literature discusses mainly the chemistry of inorganic forms. This is unfortunate because most of the metal ions in wastewater and sludge probably occur in complexed form.

Most metals are less mobile under aerobic and basic soil conditions than they are under anaerobic and acid conditions (72). The neutral water extract of soils contains less heavy metals than acidified extracts of the same soils, indicating the importance of the pH factor in influencing the mobility of metals (533). Lindsay (374) cites evidence that for zinc and copper there is a 100-fold increase in ionic activity for each unit decrease in soil pH. Normally insoluble metals may become mobilized in the event of a change in the characteristics of percolating water, such that acidic or anaerobic conditions occur.

The removal of metals by ion exchange or adsorption depends upon the availability of exchange and adsorption sites in the soil as well as on the factors (ionic activity and pH) just discussed. Therefore, the clay content of the soil is important; clay soils provide more exchange and adsorption capacity than sands and gravels (552). A correlation also exists between the form in which the elements occur in solution and their removal by exchange or adsorption. In general, elements that occur in solution as anions or neutral molecules pass through soils more readily than do elements that occur as cations. Inorganic arsenic, selenium, and fluorine in aerated soils occur as anions or neutral molecules. Although there are exceptions (depending upon the chemistry of the system), inorganic cadmium, copper, chromium, lead, mercury, silver, and zinc most commonly occur in fresh waters and soil solutions in the inorganic form as cations (615).

Only one reference was located that provided experimental data on cadmium in relation to wastewater applied to land. At Flushing Meadows, Bouwer (73) found that cadmium in wastewater applied to land in shallow basins showed very little change due to migration through the soil. The cadmium concentration dropped only slightly from 7.7 $\mu g/\ell$ to 7.2 $\mu g/\ell$. Aerobic conditions and alkaline pH prevailed in the soil studied. A study (615) by the California State Water Resources Control Board (CSWRCB) reviewed available data on soil reduction of cadmium in effluents, concluding that information is not sufficiently detailed to allow adequate evaluation of cadmium concentrations in water reaching groundwater basins. According to the study, it must be demonstrated that a particular soil is able to reduce the concentration of cadmium to a level that is acceptable for drinking water. Otherwise, the study advised against the use of wastewater effluents with concentrations above this limit for groundwater recharge operations involving percolation through soil.

Bouwer's work at Flushing Meadows (73) found that the copper concentration of applied wastewater was reduced about 86 percent by passage through the soil. This removal occurred rapidly, usually in the first 30 ft of downward flow.

Iron and manganese in well-oxidized soils are characterized by the formation of highly insoluble oxides and hydroxides. However, at low pH and under reducing conditions, these metals can be solubilized and become mobile in the soil as Fe^{2+} nd Mn^{2+}. Amramy (15) conducted a study of sewage lagoon effluent spreading on sand dunes. He found that after a subsurface travel distance of 8 m, the concentration of iron in the wastewater actually increased from 0.28 mg/ℓ to 0.57 mg/ℓ. The manganese concentration increased from 0.08 mg/ℓ to 0.19 mg/ℓ after 25 ft of travel through sand. Wesner and Baier (685) found a similar phenomenon when tracing the underground movement of wastewater after injection. Over a travel distance of 400 or 500 ft, the concentration of iron did not change. Manganese concentrations, on the other hand, increased up to 300 percent in the first 100 ft of subsurface travel. Anaerobic conditions, favoring formation of soluble manganese compounds, are mentioned as the possible cause of the large increase. These conditions are most likely caused by biological oxidation depleting oxygen near the point of injection, thus causing the reduction of manganese to soluble forms. However, after greater travel distances and the return to aerobic conditions, the manganese may revert to insoluble compounds and be removed from the migrating water.

Ragone, Vecchioli, and Ku (535) reported on deep well recharge experiments conducted in Nassau County, Long Island. Tertiary effluent was recharged by a deep well into the Magothy aquifer, the primary water supply source for Nassau County. As of September 1972, 12 recharge tests had been run since the inception of the recharge program in September 1968. Although the iron concentrations of reclaimed and native water averaged 0.44 mg/ℓ and 0.24 mg/ℓ, respectively, the iron concentration of the mixed (native and reclaimed) water at times exceeded 3 mg/ℓ. The authors mentioned several sources that could account for the increase in iron concentration, but the most probable source was the pyrite native to the Magothy aquifer. During recharge, the natural reducing condition in the aquifer was replaced by a progressively more oxidizing environment. The initial response to this change was the oxidation of pyrite, which released Fe^{+2}, SO_4^{-2}, and H^+ into solution. Eventually, ferric hydroxide precipitated, and the Fe^{+2} concentration decreased. The exact oxidation mechanism apparently involved inorganic and/or organic constituents in the reclaimed water, because water from the public potable water supply system caused no increase in iron concentration when injected into the aquifer.

The Flushing Meadows study by Bouwer (73) contained data on the reduction of lead by soil infiltration. Bouwer found that the wastewater concentration of lead decreased by 20 percent after significant travel distance underground. Apparently, a small portion of the lead was tied up rapidly (within 50 ft), while the majority was unaffected by further travel. It should be noted, however, that the soil examined in these experiments was a sand with limited adsorption and exchange capacity for many trace elements.

Both mercury and zinc form insoluble compounds in soil, lowering the activity of the ions in solution so that little movement occurs. Mercury is particularly insoluble as phosphate, carbonate, or sulfide. However, under low pH conditions, the metals may become mobilized. They can also form soluble complexes that affect their mobility under certain circumstances (388). At Flushing Meadows (73), underground percolation through 100 ft of sand produced 40 percent removal of mercury and approximately 58 percent removal of zinc. Further travel produced no further removal of mercury but reduced the zinc concentration to about 20 percent of that in the applied wastewater. The final concentrations of mercury and zinc were .00014 mg/ℓ and .037 mg/ℓ, respectively.

In conclusion, the information garnered in this study of the literature is inadequate to define fully the chemical behavior of elemental contaminants in the soil and their fate as they percolate through the unsaturated zone. The CSWRCB recommended, on the basis of its literature review (615), that wastes containing concentrations of certain metals above those acceptable for drinking water supplies should not be applied to land, unless it can be demonstrated that contamination of groundwater does not occur.

BIOCIDAL CONTAMINANTS

Use of chemical pesticides in agriculture generated many studies of the potential harmful effects of these compounds on land, crops, surface water, and groundwater. However, there is little data available on biocides in municipal wastewater or on the potential dangers of groundwater contamination through land wastewater application operations. This lack of information is understandable considering the minimal role that municipal wastewater plays in transporting biocides to the land and groundwater. Biocides come in contact with the land through a number of activities, primarily by direct application to the land for pest control. Return irrigation water, and spills and wastes from pesticide manufacturing operations also bring biocides in contact with the land.

The reactions of pesticides with soil has received limited attention in the literature. Volatilization, chemical degradation, and absorption by plant roots and seeds apparently remove a small portion of pesticides reaching the soil. A more significant process for pesticide removal may be microbiological degradation. Although this process is very slow in some cases, often taking several years, microbiological degradation accounts for the breakdown of a remarkable variety of organic compounds. Pesticides that are not removed from the soil column or broken down by these processes may be available for leaching into groundwaters.

Gerakis and Sficas (239) reviewed the literature on pesticide degradation and leaching. They reported that the most important factors involved in these two processes are soil temperature and moisture, organic matter and clay content, soil management practices, pH, and species and population density of microorganisms present. The presence of organic matter and clay in the soil appears to be positively correlated with adsorption of pesticides onto soil particles.

Van Bladel and Moreale (658) studied herbicide adsorption onto clay minerals. They found that adsorption increased with the polarizing power of the exchangeable cation, and concluded that adsorption appears to be one of the most important factors in reducing pesticide removal from soil layers by leaching.

Gerakis and Sficas (239) cite evidence that pesticides differ in their mobilities in the same soil. One study showed that (1) acidic compounds are relatively mobile, (2) phenyl ureas and triazines are of intermediate to low mobility, and (3) organochlorine compounds and organic cations are least mobile. Further data that were reviewed supported the conclusion that under normal agricultural practices and rainfall, it is very unlikely that pesticides may be leached deeply enough and in such quantities as to cause appreciable contamination of groundwaters.

A California State Water Resources Control Board report (615) mentioned that pesticides are adsorbed by soil clays, iron aluminum oxides, and especially by organic colloids, and that they are susceptible to microbial decomposition. However, the amount of biocides in average municipal wastewater was found to be so minimal that the spreading of municipal waste on land offers extremely low potential for groundwater biocide contamination.

SYNTHETIC/ORGANIC CONTAMINANTS

One of the most intensely debated questions regarding land application for treatment and/or disposal of municipal wastewater concerns the problem of residual organic contaminants. Refractory organic compounds may survive conventional treatment processes and penetrate through the soil to contaminate groundwater supplies. The controversy centers around the fate of residual organics within the soil systems, including such issues as the synergistic effects between organics and inorganics or other groundwater and soil constituents, or conversion of safe organics to hazardous compounds in the soil. Despite this controversy, no literature was found concerning groundwater pollution by the synthetic/organic contaminants in municipal wastewater as a result of land application.

The absence of literature concerning the movement through soil of synthetic/organic contaminants is not surprising, since the specific organic makeup of wastewater is unknown. Some of the chemicals of concern (PCB's, polycyclic aromatics, and other chlorinated hydrocarbons, etc.) have low solubilities in water in comparison with vapor pressures. As a result, there is a distinct possibility of vaporization when wastewaters containing these chemicals are applied to the land surface.

The California State Water Resources Control Board (615) cited a study carried out in Colorado that compared the nature of the soluble organic material in the soil profiles under a feedlot and under grassland with selected ground, well, and river waters. It was concluded that the major portion of the soluble materials in all the waters was polymeric. The soluble organics under grassland were essentially the same as those under the feedlot, although phenols were present in greater abundance in the manure and surface soil of the feedlots. About 13 percent of the soluble material in the soil profiles was carbohydrate (polysaccharides), and much of the remainder, based on IR spectra and reductive degradation procedures, appeared to be polymerized aromatic structures. This report by the CSWRCB interpreted these observations to indicate that the soluble organics under wastewater-treated soils would be similar to those under feedlot manure or grassland.

BIOLOGICAL CONTAMINANTS

Most available data suggest that virus, bacteria, and other biological pathogens present in wastewater are removed or inactivated by percolation through soil.

The California State Water Resources Control Board study (615) provides a summary of the fate of viruses, bacteria, protozoa, and parasitic worms in wastewaters applied to land. The summary states that most of these pathogens prefer warm-blooded animals as their habitat and do not flourish in the soil environment. When introduced into soils, the pathogens do not compete well with the vast number and variety of normal soil inhabitants and are subject to attack by antagonistic soil species. The time necessary for their ultimate destruction varies, according to species and environmental conditions. A compilation of pathogen survival data in the literature is shown in Table 52 below.

TABLE 52. SURVIVAL OF PATHOGENS IN SOILS (615)

Ascaris lumbricoides ova	2.5 - 7 years
Entamoeba histolytica cysts	8 days
Salmonella species	6 hours
Coliform group organisms	133 - 147 days
Q-fever organisms	148 days
Brucella abortus	30 - 100 days
Tuberculosis bacteria	6 months
Enteroviruses	12 days

The most persistent pathogens in soils appear to be ova, cysts, and spore-forming bacteria. The survival of enteric viruses in soil has not been thoroughly studied. The dependence of enteric viruses on specific host organisms for reproduction suggests that they would not multiply and would not be expected to survive for a long period of time, although survival may, at times, be long enough to cause public health concern.

Two-and-a-half years of continuous observation was conducted of wastewater reclamation by landspreading in Lodi, California (553). It was found that the MPN of coliform group organisms, which averaged 1.9 x 10^8/100 mℓ in the wastewater, was consistently reduced to less than 1/100 mℓ after 4 to 7 ft of soil travel. The average percolation rate was 0.3 ft/day in coarse-textured Hanford sandy loam. It was observed that the number of coliforms penetrating 1 ft or more was essentially independent of the coliform concentration of the wastewater.

At Whittier Narrows (441), percolation tests showed that vertical percolation of wastewater through 4 to 7 ft of soil is an effective method of removing bacteria of fecal origin, despite heavy growth of coliforms of soil origin. The formation of an organic-microbial slime layer at the water-soil interface was found to increase the efficiency of the filtering action.

Results from studies at Flushing Meadows, Arizona (73), show that fecal coliform density was reduced significantly in the first 2 or 3 ft of travel. Bouwer found that fecal coliform density at a particular depth tended to decrease with increased flooding time. The peak bacteria density invariably appeared immediately after flooding was resumed. The concentration of fecal coliforms was consistently decreased to less than 10/100 mℓ after 100 ft, and to 0/100 mℓ after 300 ft of travel.

A project at Santee, California (569), is famous for its pioneering work in the reclamation of domestic sewage for recreational lakes. Travel of secondary effluent through 1,500 ft of very coarse sand was sufficient to remove all fecal coliforms. Sampling showed that most of the coliforms were removed in the first 200 ft.

At Orange County, California (685), tests conducted on a well injection system showed coliform organisms 30 m from the injection well, but none approximately 80 m from the well. The results indicated that fecal coliforms are more easily removed by underground travel than other coliforms. Some of the other coliforms may have been supported by nutrients in the effluent.

Results of percolation tests at Lake George, New York (535), showed again that percolation of applied secondary effluent through 5 to 10 ft of soil in two different beds was sufficient to remove essentially all coliform organisms.

Browning and Mankin (83) reported an unusual case of disease outbreak due to contamination of groundwater well supplies by land application of treated sewage. In Madera, California, undisinfected secondary effluent was used to irrigate a pasture located adjacent to a deep well drawing part of the city water supply. The wastewater migrated through gopher holes, filling a construction pit around the well, and eventually flowed into the well itself.

On the basis of experience and results of full-scale, long-term wastewater reclamation studies, the CSWRCB (615) concluded that, although soil is an excellent media for removing bacteria, a small fraction of the fecal coliform bacteria therein may reach groundwater reservoirs at high percolation rates. Horizontal travel of viable fecal coliform bacteria in the aquifer does not appear to occur to a significant degree. The available data on horizontal travel, however, are inconclusive. Further investigation of the transport and survival of pathogenic bacteria in groundwater, therefore, is required.

Research shows that travel through soil removes significant amounts of viruses, primarily through adsorption. Adsorption is influenced by the pH and ionic strength of the soil solution. The available information indicates that adsorption of virus by soil is nearly complete at pH 7 or less, but decreases as the pH value increases above 7. This is mainly because the overall electric charge surrounding both the virus and soil particles becomes increasingly negative as pH levels increase and, therefore, mutual repulsion occurs (615).

It also appears that increasing cationic strength of the percolating water or soil solution increases virus removal. At the pH values normally encountered in wastewater, viruses are slightly negatively charged. The presence of calcium, magnesium, sodium, aluminum, and other positive ions in the soil solution decreases the potential for negatively charged soil and virus particles to repel each other. This results in the formation of soil-cation-virus bridges that immobilize virions (615).

The ionic strength in percolating wastewater is usually sufficient so that it does not limit adsorption. In circumstances where ionic strength is significantly decreased, however, desorption of adsorbed viruses may occur. Organic matter in wastewater can also compete with viruses for adsorption sites. In laboratory studies, when virus-adsorbed clay particles were washed with distilled water, an essentially complete desorption and reactivation of viruses took place. In field conditions, other mechanisms in soil systems may inactivate or destroy adsorbed viruses before they are subject to desorption (615). In addition to pH and ionic strength, the clay and organic matter content of soil evidently influences adsorption to some degree. In general, soils of higher clay and/or organic matter content are more effective in adsorbing viruses (615).

Definitive work on virus interaction with soil was conducted at Santee, California, where extensive studies showed that percolation through several hundred feet of soil consistently removed all virus from secondary effluent (569).

Other studies also supported the conclusion that soil effectively removes viruses. Viral analyses in Hawaii by Dugan et al. (185) showed that test soils in 5-ft lysimeters were completely effective in removing viruses. Brief tests at Whittier Narrows, California (441), achieved complete removals of Sabin Type III polio-virus vaccine. Although 250 plaque-forming units (PFU) of enteric viruses/ℓ were present in the applied wastewater, no measurable concentrations were found below 2 ft in the percolate.

In 1974 at Flushing Meadows (73), virus analyses were performed bimonthly to determine the fate of viruses in the soil system. Secondary effluent was allowed to infiltrate into six parallel horizontal basins consisting of 60 to 90 cm of fine loamy sand underlain by several coarse sand and gravel layers to a depth of 75 m, where a clay layer begins. Observation wells were installed in line across the basin area. No viruses were detected in any of the wells at any time during each flooding period. Gilbert et al. (247) stated that the failure to detect viruses in the wells indicates that the virus count was reduced by at least 99.99 percent within 3 to 9 m of basin soil.

Romero (553) reviewed the studies performed for the Department of the Army on sands ranging in classification from silty sand to coarse-granite alluvium. Results indicated that the bacterophages T1, T2, and 65 are more effectively retained in the finer sands, particularly in those containing a relatively high percentage of clay and silt. Virus removal was shown to increase with decreasing particle size. The greatest percentage of removal took place in the uppermost portion of the sand columns tested. It was shown that for a well-sorted sand of particle size averaging 0.12 mm, the removal efficiency in 2 ft of penetration was 99.999 percent.

Young and Burbank (708) described studies of virus removal in Hawaiian soils. In the laboratory, columns of various types of Hawaiian soil were subjected intermittently to percolating water with a known concentration of virus (coliphage $T4B^{II}$ mutant, and polio virus Type II (Lansing) H8), simulating the action of cesspool leaching. The effluent from each soil column was analyzed for viral content. Coliphage T4 was applied to slightly acid soils (pH 5-6) at a concentration of 2.5 x 10^6/mℓ. Percolation through 2.5 to 6 in of soil was 100 percent effective in retention of the virus. Slightly alkaline soil was less effective, removing only 67 percent of applied coliphage and 35 percent of applied polio virus in 15 in. Removal of polio virus Type II was less complete; 6-in columns were able to effect only 99.3 percent removal with an initial feed concentration of 1.5 x 10^5 pfu/mℓ.

Wellings et al. (683) found that virus can be isolated at the 6.5 m level below a spray irrigation field. Another study by Wellings et al. (682) measured virus migration through the ground from chlorinated packaged plant effluent applied to a cypress dome. Both horizontal and vertical migration was detected at distances of approximately 7 m for polio and coxsackie viruses. Wells beyond that distance showed no virus. The survival of virus within the dome was at least 28 days.

At Fort Devins, Massachusetts, where a land application site has been in operation for over 30 years, Schaub et al. (565) studied the removal of bacteria from unchlorinated primary effluent applied to soil cells. Using tracer f2 bacteriophage and the enteroviruses poliovirus I, and EMC virus it was demonstrated that tracer bacteriophage penetrated into the groundwater along with the percolating wastewater. The concentration in the groundwater stabilized at almost 50 percent of the applied virus concentration. The tracer and enteroviruses were sporadically detected at horizontal distances up to 600 ft from the application point.

Lance et al. (369) passed secondary sewage effluent containing 3 x 10^4 pfu/mℓ polio virus Type I (LSc) through 250 cm-long columns packed with calcareous sand from an area in the Salt River bed used for groundwater recharge of secondary sewage effluent. Viruses were not detected in 1-mℓ samples extracted from columns below the 160-cm level, but were detected in 5 of 43 100-mℓ samples of the column drainage water. Most of the viruses were adsorbed in the top 5 cm of soil. Virus removal was not affected by the infiltration rate, which varied between

15 and 55 cm/day. Flooding a column continuously for 27 days did not saturate the top few centimeters of soil with viruses and did not seem to affect virus movement. Flooding with deionized water caused virus desorption from the soil and increased virus movement through the columns. Drying the soil for one day between applying the virus and flooding with deionized water greatly reduced desorption, and drying for five days totally prevented desorption. The investigators concluded that large reductions (99.99 percent or more) of virus are expected after passage of secondary effluent through 250 cm or more of calcareous sand, unless heavy rains fall within one day after application of sewage.

SECTION 8

FRESH SURFACE WATER

INTRODUCTION

Approximately two-thirds of the water supplies in the U.S. are drawn from surface waters. Direct discharge of treated wastewater to these fresh surface waters is the most popular method of wastewater disposal and the most significant pathway for wastewater contaminants to reach potable water supply systems. In addition, relatively minor quantities of wastewater contaminants may indirectly reach fresh surface waters through runoff or percolation from land disposal of wastewater effluents. This section of the report discusses current knowledge about the fate of various effluent contaminants in fresh-water systems.

Most major river systems in the United States contain wastewater effluent from upstream municipalities and industries, the percentage of effluent wastewater volume varying from negligible to over 10 percent. Potable water systems utilizing these rivers as a source supply are, of course, reusing wastewater. Therefore, there is intense interest in the subject of contaminant changes which may occur in the fresh water system between waste discharge points and water intake locations.

Much of the material contained in this chapter was derived from the following references: 162, 211, 239, 326, 534, 591, and 624.

WATER QUALITY PARAMETERS

General water quality parameters in surface waters are not of direct public health concern, although they often degrade the quality of the aquatic habitat. For example, phosphorus concentrations resulting from sewage disposal contributes to luxuriant growths of certain algae, such an Anabaena, Nodularia, or Nostoc - all of which produce toxins that can be harmful to humans (618, 680). However, the tastes and odor also associated with such water degradation would generally make the water unpotable long before the concentration of toxins reached levels harmful to public health.

Suspended solids in wastewater can carry adsorbed viral and other biological contaminants (681). Trace metals occur in higher concentrations when associated with suspended matter than when they are in a dissolved state (123), a phenomenon that will be discussed more fully in the following section on elemental contaminants. Suspended matter and dissolved organics from wastewater can affect the natural biological community which, in turn, affects the nitrogen interconversions and nitrate concentrations.

The nitrogen-containing compounds (ammonia, nitrite, and nitrate) are theoretically hazardous because of nitrate's association with methemoglobinemia. Ammonia and nitrite nitrogen can be readily converted into nitrate by chemical or biological reactions. There are, however, no reported cases of detrimental public health effects resulting from nitrates in surface waters. This is because the dilution and natural processes occurring in surface waters prevent nitrate concentrations from reaching health impairing levels. There are, however, several reported cases of groundwater nitrate contamination rising to dangerous levels. These cases are discussed in the land/groundwater section of this report.

ELEMENTAL CONTAMINANTS

The behavior of elemental contaminants in fresh-water systems is very complex. Generally, elemental contaminant transport mechanisms can be divided into either elements in solution or elements associated with inorganic or biological particulates. Each of these mechanisms can be broken down still further. Dissolved elements may occur as unassociated ions or as inorganic or organic complexes. Elementals/inorganic particulate associations include coulombic attraction, as in conventional adsorption; ionic bonding, as in ion exchange; precipitated or coprecipitated metal coating; or incorporation into particulate crystalline lattices. Elementals/biological particulate associations include surface adsorption, ingested particulation, and biochemical incorporation into the organism. The particular transport mechanism that will predominate in a given water system depends, in part, on the geohydrologic environment, mineralogy/petrology of the river or lake bed, pH, temperature, dissolved organic or oxygen content, biological activity, elemental type and source, and nonelemental chemical composition of the water.

This variety of factors does much to explain the seeming discrepancies in the work of different researchers attempting to establish element distributions in fresh-water systems. For instance, Gibbs (246), in his examination of the Yukon and Amazon Rivers, concluded that precipitated metal coating and crystalline incorporation accounted for approximately 90 percent of the transported iron, nickel, copper, chromium, cobalt, and manganese (Table 53). Perhac (512), on the other hand, in his analysis of two Tennessee streams, concluded that 95 percent of the total stream content of cadmium, cobalt, copper, nickel, lead, and zinc was in the dissolved state (Table 54). Assuming that there was no gross experimental error, widely differing environmental factors must have prevailed.

The complexity of the chemistry, biology, and physics involved in water behavior of elements precludes a detailed discussion. Instead, a brief discussion is presented of the more important aspects of the behavior of elements in water, followed by a detailed examination of a few sample elements (mercury, arsenic, lead, cadmium, copper, iron) to demonstrate the principles involved.

Dissolved elements may occur as unassociated ions or as inorganic or organic complexes. Of the major elements under discussion in this report, only barium appears to any great extent as the unassociated cation; barium ions do not hydrolyze and form only weak complexes. However, several of the elements occur as unassociated anions: antimony, arsenic, boron, chromium, molybdenum, and selenium generally occur in fresh-water systems as the oxo anion. This is largely because the major sources of these elements, including wastewater, are rich in the anionic forms, and because the cationic forms are easily oxidized to the oxo anion in aquatic systems.

The majority of the dissolved elements normally exist as inorganic or organic complexes. Table 55 lists the more common ligands and the conditions and elements normally associated with them. In relatively pure water, aquo (H_2O) or hydroxo (OH^-) complexes are formed. At pH levels above neutral, many of the metal-hydroxo complexes are converted to metal hydroxides or oxides, which will precipitate out of solution or behave as colloids.

The other inorganic ligands responsible for keeping metals in solution in natural waters include carbonate, halides (notably chloride and fluoride), sulfur species (SH^-, sulfate, and sulfite), and nitrogen species (ammonia, nitrate, and nitrite). Most of the complexes formed from these ligands are thermodynamically unstable and appear as transition states between the free metal ion and a precipitate. The complexes, however, serve to keep the metals in solution for a time, and play a role in dissolving otherwise insoluble metals from precipitates or crystalline lattices.

Of somewhat more importance in terms of complex stability are the multidentate organic ligands. One of the reasons for

TABLE 53. PERCENTAGES OF THE TOTAL AMOUNTS OF IRON, NICKEL, COBALT, CHROMIUM, COPPER, AND MANGANESE TRANSPORTED BY FIVE MECHANISMS IN THE YUKON AND AMAZON RIVERS (246)

Mechanism	Iron	Nickel	Cobalt	Chromium	Copper	Manganese
	Amazon River					
In solution and organic complexes	0.7	2.7	1.6	10.4	6.9	17.3
Adsorbed	0.02	2.7	8.0	3.5	4.9	0.7
Precipitated and coprecipitated	47.2	44.1	27.3	2.9	8.1	50
In organic solids	6.5	12.7	19.3	7.6	5.8	4.7
In crystalline sediments	45.5	37.7	43.9	75.6	74.3	27.2
	Yukon River					
In solution and organic complexes	0.05	2.2	1.7	12.6	3.3	10.1
Adsorbed	0.01	3.1	4.7	2.3	2.3	0.5
Precipitated and coprecipitated	40.6	47.8	29.2	7.2	3.8	45.7
In organic solids	11.0	16.0	12.9	13.2	3.3	6.6
In crystalline sediments	48.2	31.0	51.4	64.5	87.3	37.1

TABLE 54. HEAVY METAL DISTRIBUTION IN STREAMS (512)

		Percentage of element occurring in dissolved and particulate solids			
		Sample 1	Sample 2	Sample 3	Sample 4
Cadmium	Dissolved solid	95.4	95.3	95.8	85.1
	Coarse particulate	3.9	4.2	3.5	8.9
	Colloid	0.8	0.5	0.7	6.0
Cobalt	Dissolved solid	95.9	93.2	95.9	82.3
	Coarse particulate	3.9	6.2	3.5	17.5
	Colloid	0.2	0.6	0.7	0.2
Copper	Dissolved solid	95.0	94.4	90.4	93.0
	Coarse particulate	3.6	3.8	8.2	5.8
	Colloid	1.4	1.8	1.4	1.2
Iron	Dissolved solid	18.8	12.5	26.9	20.4
	Coarse particulate	79.5	86.0	67.1	75.5
	Colloid	1.7	1.4	6.0	4.1
Manganese	Dissolved solid	23.2	41.7	18.5	10.4
	Coarse particulate	76.4	57.6	74.8	89.5
	Colloid	0.4	0.7	6.7	0.1
Nickel	Dissolved solid	96.5	95.2	96.6	84.7
	Coarse particulate	3.4	4.5	2.4	14.8
	Colloid	0.2	0.3	1.0	0.5

TABLE 54 (continued)

		Percentage of element occurring in dissolved and particulate solids			
		Sample 1	Sample 2	Sample 3	Sample 4
Lead	Dissolved solid	95.0	90.9	87.9	89.5
	Coarse particulate	5.0	7.6	9.3	10.1
	Colloid	Tr	1.6	2.9	0.4
Zinc	Dissolved solid	85.0	53.3	91.9	81.1
	Coarse particulate	15.0	46.2	7.3	18.5
	Colloid	Tr	0.5	0.9	0.4

TABLE 55. METALS COORDINATED BY LIGANDS NORMALLY FOUND IN NATURAL WATERS (521)

Metal Carried	Ligands Responsible											
	H_2O	O^{-2} or OH^-	S^{-2} or SH^-	SO_4^{-2}	SO_3^{-2}	F^-	Cl^-	CO_3^{-2}	Organic	NH_3	NH_2OH	NO_2
Boron		1										
Aluminum	2	6				1	4	6	1			6
Barium	5			P				P(c)				
Chromium	2				6		4	6(c)	1	6		6
Copper	3		6				4(b)	6(c)	1	6	(e)	6
Cobalt	4		6				4	6(c)	1	6		6
Molybdenum		1	6					6				
Manganese	3						4	6(c)	1	6		
Iron	3		6		6	1	4	6(c)	1	6(d)		
Nickel	3		6				4	6(c)	1	6	(e)	
Zinc	3		6		6		4	6(c)	1	6	(e)	
Cadmium	4						4	6	1	6	1	6
Mercury	4(a)		6		6		1	6	1			
Lead	2			P			4	6(c)				6
Arsenic		1	6				4					
Antimony		1	6				4					

Footnotes

1 In normal natural waters (pH 0-11) this metal is coordinated by this ligand.

2 Here coordination occurs only at pH of less than about 4.

3 Normally coordination only occurs at a pH of less than 4, but if the pCO_2 is low (high concentration), then bicarbonate which is water coordinated will be formed.

4 Coordination occurs at a pH of 7 or less.

5 The solubility falls markedly in the presence of this ligand at above pH due to precipitation of a carbonate or similar basic compound.

6 Coordination occurs only at pH above 7 due to ligand instability, etc.

7 Coordination occurs only at pH above 8-9.

P Precipitation almost always occurs.

(a) Water will only coordinate if no other stronger ligand is present. In some cases, there is an equilibrium.

(b) Bromide and iodide resemble chloride except that they both precipitate silver, whereas silver chloride is fairly soluble due to $AgCl_2^-$-ions at high chloride concentrations. Iodide also precipitates copper and gold.

(c) Bicarbonate usually forms carbonato complexes, but metals so marked have a soluble bicarbonate which is water coordinated. Be and Tl have soluble water coordinated carbonates, and Ag has both sparingly soluble water coordinated carbonate and hydroxide.

(d) Two valent iron in absence of air only.

(e) If ammonia is absent, a complex may be formed.

If nothing is marked, there is no coordination of this metal by this ligand in natural waters.

the lack of inorganic complex stability is that most of the inorganic ligands are monodentate, i.e., there is one ligand for each metal ion coordination site. Organic ligands are multidentate, i.e., a given ligand can usually bond to two or more of a given ion's coordination sites. A multidentate complex (chelate) is more stable than a corresponding complex with monodentate ligands; thus, a chelate complex is apt to keep a metal ion in solution far longer than will an inorganic complex. Normally, this is not a problem; in relatively unpolluted fresh water the organic content is low, but in highly polluted water unusually high soluble metal concentrations may result. Furthermore, there is evidence that some synthetic organic ligands, such as may be found in wastewater, form stronger complexes than do natural organic ligands (384). It has been demonstrated that one synthetic ligand, nitrilotriacetate (a proposed substitute for phosphates in detergents), is capable of dissolving significant quantities of precipitated lead out of bottom sediments (268, 715).

The tendency for a given metal ion and ligand to form complexes depends on solution pH, concentration of the metal and ligand, concentrations of other metals and ligands in solution, equilibrium constants, redox conditions, and so forth. No two elemental contaminants behave exactly alike, and even different oxidation states of the same element may exhibit widely varying solution chemistry. Moreover, natural water systems are seldom in an equilibrium condition before, much less after, wastewater addition. This constantly changing system makes concise pathways almost impossible to construct. In general, low pH, an oxidizing environment, and the presence of a variety of ligands enhance element solution tendencies. This is significant from a public health standpoint, because soluble elemental species are much more readily available for human contact than are precipitates or particulate elementals.

Elemental/inorganic particulate interactions typically account for the bulk of the nondissolved elemental fraction. These interactions include coulombic or ionic attraction, precipitated or coprecipitated coating, or lattice incorporation. Coulombic attraction, or adsorption, is the least important transport mechanism except in the case of colloidal particulates, such as microparticulate iron or manganese hydroxides, which carry a weak negative charge and attract elemental cations. Larger particulates do not possess a strong enough charge to make coulombic attraction important.

Ionic attraction, or ion exchange, is somewhat more important. In this process, the heavy metal elements (Mn, Fe, Co, Ni, Cu, Zn, Mo, Cd, etc.) replace alkali and alkali earth cations (K, Na, Li, Mg, Ca) attached to crystalline lattices by ionic bonds. These ionic bonds will hold unless (1) the element is displaced by another element forming a stronger ionic bond, (2) a ligand forming a coordination bond stronger than the ionic bond breaks the ionic bond, or (3) an excess of alkali or alkali earth cations is available to force the equilibrium back to its original state.

According to Gibbs (246), lattice incorporation is generally insignificant as a transport or removal mechanism of dissolved wastewater elements. It is a slow process that takes place in the sediments and has a more significant impact on the elemental composition of the sediments. The lattice-incorporated element burden of a water system is primarily from weathered rock, sand, and clay.

Precipitated and coprecipitated metal coatings account for most of the non-native particulate element content of a water system. Under favorable pH-EH conditions, metal precipitates will form. The initially small precipitate particles will tend to agglomerate or adhere to any available surface. In addition, cations held to particulates by coulombic or ionic bonds can form covalent bonds with anionic components of the particulate, if solution redox potentials change. In either case, the particulates are left with a coating of metal precipitates. This coating ultimately settles out of solution with the particulate, removing the elements from possible ready human contact unless they are redissolved by a change in redox conditions, a strong ligand, or some other agent strong enough to attach the precipitate. These coatings may account for the bulk of the inorganic particulate element burden of wastewaters.

Elemental contaminants incorporated with biological particulates constitute the remainder of the particulate elemental burden of a water body or wastewater. This incorporation may take the form of surface adsorption, inorganic particulate ingestion, or biochemical incorporation into an organism's tissues. Obviously, the role played by biological transport is highly dependent on the type and quantity of organisms present. Surface adsorption is usually associated with microorganisms. Particulate ingestion is associated with those organisms that have internal digestive organs (e.g., fish, crustaceans, worms, etc.); ingested particulates are usually eliminated within a short time, but the elements associated with them may be biochemically incorporated into the organism's tissues. From a public health standpoint, soluble and biochemically incorporated elementals are the most important, for it is by these routes that potentially hazardous elements reach man. Biochemical incorporation involves both essential trace element concentration (e.g., cobalt in vitamin B-12) and reaction of an element with cellular chemicals (e.g., the reaction of mercury with sulfur-containing amino acids in proteins). Both plants and animals are involved and, thus, the concentration of a given element may move up the food chain. The incorporation is reversible; once the organism is removed from contact with the element, the latter can gradually be excreted.

No two elements behave exactly alike; furthermore, the number of factors available that can affect transport mechanisms makes the possibilities nearly endless. There are similarities, however, that make the use of examples illustrative and useful; mercury, arsenic, iron, cadmium, copper, and lead will be used to provide detailed descriptions of the general pathways discussed above.

Mercury from wastewater enters a water system primarily as the metal or divalent cation. Although of limited solubility, it can reach concentrations of 100 ppb in aerated water (234). Metallic mercury alone is soluble up to 25 ppb and will hydrolize to soluble $Hg(OH)_2$ in oxygenated systems, increasing the overall solubility and water content. Despite these solubility figures, mercury concentrations seldom exceed 5 ppb (Table 56) except in polluted water.

Inorganic and biological adsorption, absorption, and precipitation serve to keep the concentrations of dissolved mercury much lower than the theoretical maximum. In general, the bulk of the mercury in a given water system is in the sediments; Table 57 gives a summary of some of the mercury concentrations in the sediments of Lake Erie. In reducing sediments, mercury is tied up as the sulfide, although if the system becomes sufficiently alkaline, HgS_2^- may be released into solution. Should the sediments become aerobic, the sulfide will be oxidized to sulfate, and the mercury will be released.

All soluble mercury species except mercuric sulfide can be absorbed by bacteria. Once the mercury is in the bacteria, a series of transformations - possibly via a detoxification mechanism - convert the incorporated mercury into mono- and dimethyl mercury, both soluble at low concentrations and readily released into solution. The methyl mercury compounds are much more lipid-preferring than the inorganic forms and are quickly absorbed by living tissues. As a rule, mercury concentrations tend to increase in organisms up the food chain, so that the highest concentrations are found in fish. This is partly due to absorption of methyl mercury from the water and partly from ingestion of plants or smaller organisms containing methyl mercury. When the organisms die, the mercury returns to the sediments, where most of the bacterial methylation occurs. Table 57 lists some sample sediment and plankton/algae mercury concentrations in Lake Erie.

Arsenic, selenium, and antimony are chemically similar and exhibit analogous environmental behavior. Arsenic has been studied far more than either selenium or antimony. The following

TABLE 56. SELECTED CONCENTRATIONS OF MERCURY IN NATURAL WATERS (234)

Source and Location	Mercury (ppb)
River water, European USSR	0.4-2.8
River water, Armenia	1-3
Saale River, Germany	0.035-0.145
River water, Italy	0.01-0.05
River water, near mercury deposits, Italy	up to 136
Colorado River, Arizona	<0.1
Ohio River, Illinois	0.1
Mississippi River, Kentucky	<0.1
Missouri River, Montana	<0.1
Missouri River, St. Louis, Missouri	2.8
Kansas River, Topeka, Kansas	3.5
Hudson River, New York	0.1
Lake Champlain, New York	<0.1
Maumee River, Antwerp, Ohio	6.0
Delaware River, New York	<0.1

TABLE 57. MERCURY CONTENT OF SEDIMENTS AND PLANKTON/ALGAE SAMPLES COLLECTED FROM LAKE ERIE (519)

Station No.	Approximate Location	Mercury content in µg/g[a]	
		Sediments[b]	Plankton/Algae
01	Buffalo River	2.0	31.2
02	Cattaraugus Creek	1.2	25.1
03	Barcelona	0.6	2.8
04	Ashtabula	4.6	7.4
05	Fairport	1.5	12.8
06	Cleveland	12.0	33.5
07	Toledo	10.4	20.5
08	Detroit River	4.5	26.1
09	Mid. Bass Island	1.5	20.1
10	Port Crewe	0.5	12.4
11	Port Stanley	1.5	12.0
12	Long Point	7.0	14.7
13	Long Point Bay	1.0	23.7
14	Port Maitland	1.8	15.4
15	Mid-Lake	1.5	0.6
16	Black Rock Channel	12.4	27.8

[a] In terms of the equivalentdry wt of the sample.

[b] Sediment samples from 3 to 30 cm below the water-sediment interface.

discussion of arsenic is largely applicable to selenium and antimony as well.

Arsenic has an unusually complex chemistry in aquatic systems: oxidation-reduction, ligand exchange, precipitation, adsorption, and biomethylation reactions all take place. Arsenic species can be removed from water via surface adsorption and coprecipitation with metal ions; both arsenate (AsO_4^{-3}) and arsenite (AsO_3^{-3}) have a high affinity for hydrous iron oxides and readily coprecipitate with or adsorb onto them. Significantly, iron ores are always enriched with arsenic (214). Aluminum hydroxide and clays are adsorb arsenate species, although to a lesser degree.

Microbial transformations of arsenic, while demonstrable in the laboratory, have not been positively identified in natural water systems. The two most commonly postulated transformations are oxidation of arsenite and methylation. Methylation is important because it could be a means by which sediment arsenic is recycled back into the water system; natural aquatic methylation has not been demonstrated.

Soluble iron entering a water system in wastewater will usually be either ferrous (Fe II) or complexed ferric (Fe III) iron. The former is much more soluble than the latter (which has a stronger tendency to form complexes), although neither tends to remain in solution long. In the surface layers of most natural waters, pH levels and oxygen conditions are such that Fe (II) is readily oxidized to Fe (III), which just as readily hydrolyzes to insoluble hydrous ferric oxide (FeOOH). Hydroxide has a much stronger affinity for ferric iron than do basic organic or inorganic ligands.

Hydrous ferric oxide tends to form microcrystalline precipitates of a colloidal nature, so that it is almost impossible to analytically distinguish between soluble and colloidal iron. Consequently, the two forms of iron are usually reported together as soluble iron. Although hydroxide supersedes other anionic ligands, frequent incorporation of coordinating anions into ferric oxide precipitates enhances colloidal stability and further blurs the distinction between the colloid and soluble ferric complexes.

Cadmium readily precipitates as the hydroxide or carbonate and consequently is not normally found in high concentrations in surface waters. In fact, several researchers (188, 461) have noted that high soluble cadmium concentrations are invariably associated with polluted water that receives a steady cadmium source, such as industrial wastewater.

Cadmium (II) readily hydrolyzes and forms transitory inorganic complexes, such as chloride complexes that have a limited affinity for hydrous iron and manganese oxides, and organic particulates. The organic affinity probably indicates a reaction between the cadmium and sulfur-containing compounds.

Cadmium forms the insoluble hydroxide at pH levels of 7 and above; it forms the insoluble carbonate under oxidizing conditions, particularly in soft waters where cadmium does not have to compete with calcium and magnesium for the carbonate anion. Once cadmium has precipitated and settled into the sediments, it is not readily removed. Consequently, if cadmium additions are reduced, a water body will tend to purify itself of soluble cadmium.

Copper, and to a lesser extent nickel, occupy an unusual position in water chemistry and biology because they are both nutrients and toxins. This has a pronounced effect on their water chemistry. Copper contained in wastewater may be either soluble or particulate; neither form predominates as a rule. Copper adsorbs readily onto clay and organic particulates. Copper also forms several very stable complexes. In pure water, while the aquo complex may predominate, the carbonate, chloride, and amine inorganic complexes are much more stable.

Ultimately, the soluble stability of copper can be attributed to organic complexes, since copper forms coordination complexes with virtually every conceivable organic ligand. These complexes are very stable thermodynamically and are also resistant to microbial attack, a mechanism responsible for the destruction of most organic complexes. Copper is a bacterial toxin and, if released from its complex by microbial attack, simply kills the offending bacteria and forms a new complex (461).

Copper is removed from solution via precipitation or biological incorporation. Since the most common precipitate is the carbonate, most sediment copper is in the carbonate form (140, 461). An essential trace nutrient, copper is readily incorporated into aquatic plants and animals.

The main soluble species of lead in wastewater are the lead (II) cation and the hydrolyzed complex Pb $(OH)_3^-$. Lead forms a variety of stable complexes as well; researchers (381) have identified both $PbOH^+$ and $Pb(CO_3)_2^{-2}$ in natural water systems. Lead complexes easily with a variety of organic chelates, forming very stable complexes. Some of these complexes are more stable than the sediment lead precipitates; therefore, they will actually dissolve otherwise insoluble lead. A case in point is nitrilotriacetate, which can solubilize lead from lead carbonate precipitates (268).

Low water organic content generally prevents solution lead concentrations from exceeding a few parts per billion. In most water systems, lead introduced with wastewater readily forms insoluble $Pb(OH)_2$ and $PbCO_3$, which will precipitate and adsorb onto suspended particulates. Ionic lead is not so strongly adsorbed, although it does have some affinity for clays.

Hydrous iron oxides strongly sorb ionic lead at neutral to slightly acidic pH levels. Some ionic or complexed lead adsorbs onto or is chelated with the surface mucilage of algae, and microorganisms immobilize substantial quantities of inorganic lead, presumably on or in all membranes (648). As a result of all of these mechanisms, most of the water lead burden is associated with particulate matter, and most of the lead entering a normal water system ultimately finds its way into the sediment.

Natural water bodies normally contain very low dissolved concentrations of the more harmful elemental contaminants. Unless wastewater additions are voluminous and repeated, natural water chemistry can purify the water of soluble species fairly well. However, there is a buildup of these elements in the sediments. This means that if the local water chemistry should change significantly, the elements can still be released to solution. Natural water purification mechanisms can only change the wastewater element problem from a real to a potential hazard; they cannot solve the problem of element contamination.

BIOCIDAL CONTAMINANTS

In general, municipal wastewater will have detectable quantities of biocides only if it contains biocide manufacturing wastes. The single most important source of biocidal contaminants in fresh-water bodies is surface runoff, followed by aerial fallout and industrial waste discharge from plants manufacturing biocides. Cleanup and disposal by households, farmers, gardeners, etc., contribute minimally to the overall wastewater burden.

In this discussion, biocides will be classed as chlorinated hydrocarbons, organophosphates, carbamates, and ionic biocides (Table 58).

Biocides can be transported or removed from the system by microbial or chemical degradation, photodegradation, adsorption to sediment or humic matter, adsorption, volatilization, and biological uptake. All of these mechanisms are in turn affected by pH, temperature, salt or organic content, and bioproductivity. One mechanism that has not been studied to any great degree - especially in fresh water - is aerosolization. This mechanism will be discussed in greater detail in the marine water section of this chapter, as its importance in fresh-water systems is limited mainly to the larger lakes. Briefly, however, aerosolization occurs through the action of wind and waves on floatables.

TABLE 58. BIOCIDE TYPES AND EXAMPLES

Chlorinated Hydrocarbons

- DDT (DDD, DDE)
- Methoxychlor
- Endrin
- Dieldrin
- Aldrin
- Toxaphene
- Lindane
- Chlordane
- Heptachlor

Organophosphates

- Parathion
- Malathion
- Dimethoate
- Methyl parathion
- Phorate
- Demeton
- Ethion
- Disulfaton

Carbamates

- Carbaryl
- Sevin
- Baygon
- Pyrolan
- Dimetilan

Ionic Biocides

- Diquat
- Paraquat
- Chlormequat
- Morfaunquat
- Phosphon
- Hyamine
- 2,4-D
- 2,4,5-T
- Dalapon
- Silvex
- Dichlobenil

Aerosols and particulates can be released to the air and transported great distances by the wind; biocides can concentrate in floatables via dissolution in surface oil, adsorption on floating matter, or flotation (caused by low specific gravity and insolubility), thus becoming amenable to the aerosolization process. As mentioned above, this is not an important transport mechanism in most fresh-water systems.

Various transport mechanisms affect the biocide classes differently. This is demonstrated in Table 59, which compares the persistence of selected chlorinated hydrocarbons, organophosphates, and carbamates in river water. Because of these differences, the biocide classes will be discussed separately.

The chlorinated hydrocarbon pesticides are all insoluble in water, with the exception of lindane, which is sparingly soluble to 10 ppm (211). They are generally resistant to microbial and chemical degradation, as evidenced by their estimated environmental half-lives, shown in Table 60.

TABLE 60. ESTIMATED PESTICIDE HALF-LIVES (211)

Pesticide	Half-Life, yrs
Lindane	2
Chlordane	8
Toxaphene	11
Heptachlor	2 to 4
DDT	10 to 20
Endrin (Dieldrin)	8 to 10

These pesticides are somewhat more susceptible to photodegradation, although the degradation products are often as toxic as the parent compound, regardless of the type of degradation. DDT is decomposed chemically to DDD and DDE and photochemically to PCB's (162, 550); aldrin is photooxidized to the more toxic dieldrin (162); and methoxychlor is degraded to methoxychlor DDE (501). Surface oil slicks tend to concentrate chlorinated hydrocarbons and thus make them more available for photochemical degradation (162).

Chlorinated hydrocarbons in general readily adsorb onto fungi, algae, and floc-forming bacteria (385, 501), and thus tend to concentrate in biological communities. When ingested by higher organisms, they accumulate in lipid tissues; consequently, there is a tendency for chlorinated hydrocarbons to concentrate up the food chain.

Chlorinated hydrocarbon insecticides differ in chemical structure, but they all exhibit affinity for organic sediments

TABLE 59. PERSISTENCE OF COMPOUNDS IN RIVER WATER (415)

Compound	Original compound found, percent				
	0-time	1 wk	2 wk	4 wk	8 wk
Organochlorine compounds					
BHC	100	100	100	100	100
Heptachlor	100	25	0	0	0
Adlrin	100	100	80	40	20
Heptachlor epoxide	100	100	100	100	100
Telodrin	100	25	10	0	0
Endosulfan	100	30	5	0	0
Dieldrin	100	100	100	100	100
DDE	100	100	100	100	100
DDT	100	100	100	100	100
DDD	100	100	100	100	100
Chlordane (tech.)	100	90	85	85	85
Endrin	100	100	100	100	100
Organophosphorus compounds					
Parathion	100	50	30	<5	0
Methyl parathion	80	25	10	0	0
Malathion	100	25	10	0	0
Ethion	100	90	75	50	50
Trithion	90	25	10	0	0
Fenthion	100	50	10	0	0
Dimethoate	100	100	85	75	50
Merphos	0	0	0	0	0
Merphos recov. as Def	100	50	30	10	<5
Azodrin	100	100	100	100	100
Carbamate compounds					
Sevin	90	5	0	0	0
Zectran	100	15	0	0	0
Matacil	100	60	10	0	0
Mesurol	90	0	0	0	0
Baygon	100	50	30	10	5
Monuron	80	40	30	20	0
Fenuron	80	60	20	0	0

and resistance to microbial attack. As a result, there is accumulation in bottom sediment. Research on Lake Michigan demonstrates this, as shown in Table 61. Routh (558) showed that DDT, with its affinity for fine particulate clay sediments, concentrated up to 20 times normal background levels, from 10 to 200 ppb. This affinity for organic matter and particulates leads to high sediment DDT concentrations. Table 62 shows DDT concentrations in stream sediments over a period of time. Adsorption of DDT on algae can be 10 to 100 times greater than adsorption on clay (350). Moreover, DDT seems to have an inhibitory effect on sediment bacteria (11).

There has been little research on other chlorinated hydrocarbons, much of it limited to an evaluation of environmental levels. Table 63 gives the results of one such survey for dieldrin.

TABLE 63. DIELDRIN IN RIVER BOTTOM SILTS (466)

Source	Dieldrin (ppb)
Iowa River	8.8
Des Moines River	35
East Nishnabotna River	21
West Nishnabotna River	16
Upper Iowa River	<1
Johnson County Creek	170

Other researchers have found that the highest reported concentrations of several pesticides in major U.S. river basins from 1958 to 1965 were as follows: dieldrin, 0.100 μg/ℓ; endrin, 0.116 μg/ℓ; and DDT, 0.148 μg/ℓ. Dieldrin was the most widely found pesticide (679).

The organophosphorus biocides are more soluble than the chlorinated hydrocarbons. These solubilities range from 1 ppm for ethion to 20,000 ppm for dimethoate; most fall in the 25 to 150 ppm range (211). The organophosphorus biocides are also more amenable to both microbial and chemical degradation. Even parathion, the most chemically resistant of the organophosphates, will degrade via ester linkage hydrolysis in a few months under normal conditions. The degradation takes place in just a few weeks in polluted water with a high bacteria count (264). Yu and Sanborn's (709) experimental evaluation of parathion in a model ecosystem yielded a calculated half-life of 15 to 16 days. In a similar study, guthion yielded a half-life of one month at pH levels less than 9 and a half-life of less than one week at more alkaline pH's.

TABLE 61. CHLORINATED HYDROCARBON INSECTICIDES IN SOUTHERN LAKE MICHIGAN SEDIMENTS (μg/l) (382)

	Top interval			2-6 cm			6-12 cm		
Insecticide	Median	50% Mid-range	No. of Samples	Median	50% Mid-range	No. of Samples	Median	50% Mid-range	No. of Samples
p,p'-DDT	9.3	5.5-17	59	3.8	2.6-5.2	40	3.0	1.5-6.0	20
o,p-DDT	1.2	Tr-2.0	54	0.7	Tr-1.0	37	Tr	ND-2.3	14
p,p'-DDE	2.2	0.6-3.5	59	0.8	Tr-1.5	40	0.6	Tr-2.1	19
o,p-DDE	Tr	Tr	49	ND	ND	32	ND	ND-Tr	12
p,p'-DDD	3.0	1.4-10	54	0.5	Tr-2.0	37	ND	ND	12
o,p-DDD	Tr	Tr-1.8	49	ND	ND-0.5	32	ND	ND	12
Total DDT complex	18.5	10-32	59	6.3	3.9-13	40	3.4	2.2-8.1	20
Dieldrin	2.0	1.3-4.1	54	Tr	Tr-1.1	37	Tr	Tr-0.9	14
Heptachlor epoxide	Tr	Tr-0.7	54	Tr	ND-0.5	32	ND	ND	14
Lindane	Tr	Tr	45	ND	ND-Tr	30	ND	ND	14

TABLE 62. DDT CONCENTRATIONS IN STREAM SEDIMENTS (271)

Years after one application	DDT (ppm)
0	.83 (.04-1.6)
1	1.08 (.25-1.9)
2	--
3	.21 (.12-.30)
4	.21 (.16-.25)
5	.59 (.07-1.9)
6	.06 --
7	.21 (.11-.31)
8	.13 --
9	.03 (.02-.04)
10	.07 (0-.16)
Never sprayed	.006 (0-.02)
Two sprays	.43 (.17-.77)
Three sprays	.35 (.17-.6)

Interestingly, the degradation of organophosphates can be inhibited by the presence of other synthetic organic chemicals. Experiments were conducted with two detergent surfactants - alkyl benzene sulfonate (ABS) and linear alkyl benzene sulfonate (LAS). These experiments demonstrated increased persistence for several organophosphate insecticides, especially parathion and diazinon (162). As a result, highly polluted water may exhibit accumulations or half-lives far beyond the normal for organophosphates, which, as a rule, neither persist nor accumulate in the environment, but are removed entirely within a few months.

Carbamate biocides are moderately soluble, ranging from 7 ppm for terbutol to 250 ppm for propham and averaging around 100 ppm (211). In general, they decompose easily and show little tendency toward adsorption on suspended material, but hydrolyze readily. The hydrolysis is particularly pH dependent, virtually ceasing entirely below pH 5 (211) and increasing as the pH and temperature rise. High salt content affects the hydrolysis rate inversely, slowing the rate as the salt concentration increases (211). Carbamates photodecompose readily - increasingly so, as the pH rises - and can be rapidly biodegraded under normal circumstances (550). Carbamates are not, then, persistent in normal water systems, lasting only a few days to a few weeks, but remain as a stable compound in acidic waters (211).

Ionic biocides are a broad class embracing a variety of chemical types and uses. They are all considered soluble in water, with solubilities ranging from 100 to more than 1,000,000 ppm. Ionic biocides that are marginally soluble in pure water have increased solubilities in natural waters high in humic acid salts (211). With few exceptions, these biocides do not accumulate or persist and, consequently, are seldom found in high concentrations.

Ionic biocides are, however, strongly adsorbed onto soil particles, all types of clay, humic matter, and organisms - in short, onto anything with a partial charge or an ion exchange capability (211). They are generally resistant to chemical attack but photodegrade readily, except when adsorbed onto particulate matter (211). Ionic biocides respond differently to microbial attack, but are absorbed by many organisms. As a result, they tend to concentrate in organisms and up the food chain. Research on TCDD, an ionic herbicide residue, demonstrated that accumulation was directly related to water concentrations (0.05 to 1,330 ppb) and averaged between 400,000 and 2,000,000 times the water concentration (321).

SYNTHETIC/ORGANIC CONTAMINANTS

Recently, there has been a great interest in identifying synthetic/organic trace compounds in water supplies drawn from rivers and in other water bodies receiving treated wastewater. Although studies have been made of the concentrations found in various water systems, neither the environmental pathways nor the potential health effects to man of these substances have been studied to any great extent.

Over 100 synthetic/organic compounds have been identified in various drinking water sources. Thirty-six compounds were found in the lower Tennessee River (Table 64), while 66 were identified and quantified in Mississippi River water at New Orleans (Table 65). Table 66 lists the results of organic analyses of several other domestic water supply sources.

The many different types of compounds under discussion here make generalizations difficult regarding their environmental fate. For instance, acetone is infinitely soluble in water; chloroform is soluble to about 8,200 mg/ℓ; carbon tetrachloride is soluble to about 800 mg/ℓ; and n-decane is insoluble. The specific gravity of toluene is less than that of water, while the specific gravity of carbon disulfide is greater. Acetaldehyde is readily metabolized since it is a natural metabolic intermediate, but branched alkyls are almost impervious to microbial attack.

The differences in man-made synthetic/organic compounds exceed the similarities, but in general, these compounds are persistent and resist microbial degradation. Beyond that generalization, research has been too limited to discuss specific compounds in detail.

Polychlorinated biphenyls (PCB's) are the only class of synthetic/organic contaminants that have been studied in detail. They are virtually insoluble in water, which, combined with a high specific gravity and volatility, serves to keep solution PCB concentrations low. However, PCB's are strongly adsorbed onto suspended particulate matter and transported through the water system. Because of their heavy, insoluble character and sediment affinity, they tend to accumulate in bottom sediments. A comparison of selected water and sediment PCB concentrations from across the United States is presented in Table 67.

PCB's are fairly stable in fresh-water systems, resisting hydrolysis and chemical degradation, and are not amenable to photodegradation (481). Theoretically, they should readily vaporize from solutions, but this is prevented by their tendency to sink or strongly adsorb onto suspended matter. Only PCB's that are associated with floatables or oil slicks appear to vaporize to any great degree. The lower isomers (four or fewer chlorine atoms) are somewhat responsive to biodegradation, but the degradation products are frequently more toxic than the PCB itself (481). The higher isomers resist microbial attack.

TABLE 64

ORGANIC COMPOUNDS IDENTIFIED TO DATE FROM LOWER TENNESSEE (615)

COMPOUNDS

Acenaphthene
Allylbenzoate
Anthracene
Benzene
Biphenyl
Butylbenzene
5-Chloro-2-Methylbenzofuran
p-Cresol
Diallyl Adipate
Dibutyl Phthalate
Diphenylacetylene
1,1-Diphenylethene
2,6-Di-Tert-Butyl-4-Methylphenol
Ethylbenzene
Ethyl o-Phthalate
Ethylstyrene
Ethylene Dimethylacrylate
Fluoranthene
Fluorene
Hexachlorobenzene
Indene
o-Methoxybenzoic Acid
2-Methylanthracene
2-Methylbiphenyl
4-Methyldiphenylacetylene
Methyl Indene (2 isomers)
1-Methylnaphthalene
Naphthalene
p-Nonylphenol
n-Octyl-o-Phthalate
Pyrene
Styrene
1,2-Tetradecanediol
Toluene
3,4,4-Trimethyl-2-Hexene
Xylene

TABLE 65

ORGANIC COMPOUND IDENTIFICATIONS

NEW ORLEANS AREA WATER SUPPLY STUDY

(615)

		Highest Measured Concentration μg/l		
	Compound	Carrollton Water Plant	Jefferson # 1 Water Plant	Jefferson # 2 Water Plant
1	Acetaldehyde	D-VOA	NE	NE
2	Acetone	D-VOA	NE	NE
3	Alkylbenzene-C_2 isomer	0.05	ND	ND
4	Alkylbenzene-C_2 isomer	0.33	ND	ND
5	Alkylbenzene-C_2 isomer	0.11	0.03	ND
6	Alkylbenzene-C_3 isomer	0.01	ND	ND
7	Alkylbenzene-C_3 isomer	0.04	0.05	0.02
8	Alkylbenzene-C_3 isomer	0.02	ND	ND
9	Atrazine * (2-chloro-4-ethylamino-6-isopropylamino-s-triazine)	5.0	4.7	5.1
10	Deethylatrazine (2-chloro-4-amino-6-isopropylamino-s-triazine)	0.51	0.27	0.27

TABLE 65 (continued)

Compound		Highest Measured Concentration µg/l Carrollton Water Plant	Jefferson # 1 Water Plant	Jefferson # 2 Water Plant
11	Benzyl butyl phthalate*	0.64	0.81	0.73
12	Bromodichloroethane	D-VOA	NE	NE
13	Bromoform *	0.57	ND	ND
14	Butanone	D-VOA	NE	NE
15	Carbon disulfide	D-VOA	NE	NE
16	Carbon tetrachloride	D-VOA	NE	NE
17	bis-2-Chloroethyl ether*	0.07	0.16	0.12
18	Chloroform *,a	113	NE	NE
19	bis-2-Chloroisopropyl ether *	0.18	0.05	0.03
20	n-Decane *	0.04	ND	ND
21	Decane-branched isomer	0.03	ND	ND

TABLE 65 (continued)

		Highest Measured Concentration µg/l		
	Compound	Carrollton Water Plant	Jefferson # 1 Water Plant	Jefferson # 2 Water Plant
22	Dibromodichloroethane isomer	0.33	ND	0.63
23	Dibromochloromethane *	1.1	0.30	0.60
24	Dibutyl phthalate *	0.10	0.16	0.19
25	2,6-Di-t-butyl-p-benzoquinone *	0.22	0.19	0.23
26	Dichlorobenzene isomer	0.01	D-RE	ND
27	1,2-Dichloroethane [a]	8	NE	NE
28	Dichloromethane	D-VOA	NE	NE
29	Dieldrin **	0.05	0.07	0.05
30	Diethyl phthalate *	0.03	0.03	0.01
31	Di(2-ethylhexyl) phthalate *	0.10	0.31	0.06
32	Dihexyl phthalate	0.03	ND	ND

TABLE 65 (continued)

	Compound	Highest Measured Concentration µg/l		
		Carrollton Water Plant	Jefferson # 1 Water Plant	Jefferson # 2 Water Plant
33	Dihydrocarvone	0.14	0.06	0.07
34	Diisobutyl phthalate *	0.59	ND	ND
35	Dimethyl phthalate	0.27	0.13	0.18
36	Dioctyl adipate	0.10	ND	ND
37	Dipropyl phthalate *	0.07	0.13	0.14
38	n-Dodecane *	0.01	ND	ND
39	Endrin **	0.004	NYE	NYE
40	Ethanol	D-VOA	NE	NE
41	o-Ethyltoluene *	ND	0.04	0.02
42	p-Ethyltoluene *	0.02	0.03	0.03
43	1, 2, 3, 4, 5, 7, 7-Heptachloronorbornene *	0.06	0.05	0.05

TABLE 65 (continued)

		Highest Measured Concentration µg/l		
	Compound	Carrollton Water Plant	Jefferson # 1 Water Plant	Jefferson # 2 Water Plant
44	Heptachloronorbornene isomer	0.06	0.04	0.04
45	Hexachloro-1,3-butadiene *	0.16	0.27	0.21
46	Hexachloroethane *	4.4	0.19	0.16
47	Isophorone *	1.5	2.2	2.9
48	Limonene *	0.03	ND	ND
49	Methanol	D-VOA	NE	NE
50	Methylbenzoate	ND	D-RE	ND
51	3-Methylbutanal	D-VOA	NE	NE
52	2-Methylpropanal	D-VOA	NE	NE
53	n-Nonane *	0.03	ND	ND
54	n-Pentadecane *	0.02	ND	ND

TABLE 65 (continued)

	Compound	Highest Measured Concentration µg/l Carrollton Water Plant	Jefferson # 1 Water Plant	Jefferson # 2 Water Plant
55	Tetrachloroethane isomer	0.11	ND	ND
56	Tetrachloroethylene	D	0.5	0.41
57	n-Tetradecane *	0.02	ND	ND
58	Toluene *	0.08	0.10	ND
59	1,1,2-Trichloroethane *	0.35	0.45	0.41
60	1,1,2-Trichloroethylene	D-VOA	NE	NE
61	n-Tridecane *	0.01	ND	ND
62	Trimethyl-trioxo-hexahydrotriazine isomer	0.07	ND	ND
63	Triphenyl phosphate *	0.12	ND	ND
64	n-Undecane *	0.02	ND	ND
65	Undecane-branched isomer	0.04	ND	ND
66	Undecane-branched isomer	0.06	ND	ND

KEY TO SYMBOLS USED IN TABLE 65

Symbols used in column headed "Compound"

*	While all compounds listed in the table were identified by one or more methods, those marked with this symbol gained added confirmation by gas chromatography retention time match with an available standard of the compound.
**	Compounds marked with this symbol gained further confirmation by gas chromatography retention time match with available standards on each of three different columns, polar and non-polar.
a	The quantitative values for these compounds were obtained on Volatile Organics Analysis by comparison with standards of known concentration at the Water Supply Research Laboratory. Compound 18 was detected but not quantified in Tetralin extracts of Carrollton water at Southeast Environmental Laboratory, but not in Tetralin extracts of Jefferson No. 1 or Jefferson No. 2. The latter laboratory did not detect compound 27.

Symbols used in columns headed "Highest Concentration Measured"

D-VOA	These compounds were detected by Volatile Organics Analysis - Bellar Technique only. Quantitative values have not yet been obtained. This method was performed only on the Carrollton water at the Water Supply Research Laboratory.
D-RE	These compounds were detected only on XAD resin extracts in the specific water for which this symbol is used. Quantitative values were not obtained from the resin extracts. The compound may have been detected and quantified by another method in one or both of the other waters examined.
D	In the one instance where this symbol was used the compound was detected by both the Water Supply Research Laboratory and Southeast Environmental Research Laboratory but not quantified by either laboratory.
NE	This symbol means not examined. It is used exclusively for some compounds reported by the Water Supply Research Laboratory. This laboratory did not obtain samples of water from Jefferson No. 1 or Jefferson No. 2.

KEY TO TABLE 65 (Continued)

ND	This symbol means the compound was not detected in that specific water by any of the methods employed.
NYE	Compound 39 was confirmed in Carrollton water carbon chloroform extracts shortly before preparation of this report. Jefferson No. 1 and Jefferson No. 2 extracts have not yet been re-examined specifically for compound 39.

TABLE 66. MOLECULAR CONSTITUENTS IDENTIFIED IN NATURAL WATER SAMPLES (615)

Constituent	Sample Source[a]	Concentration mg/ℓ
p-Cresol	3	7
Diethylene glycol	5	1
Ethylene glycol	5	20
Glycerine	1,2,3,4,5	1-20
Glycine	1	2
Mannitol	5	2
Methyl-a-D-glucopyranoside	4	30
Methyl-B-D-glucophranoside	4	3
Sucrose	1,5	2
Xylitol	5	1
Urea	1,2	4
Inositol	1,2,3,4,5	0.5-1
O-Methylinositol	1,2,3,4,5	0.3-10
Linoleic Acid	1,5	1
Oleic Acid	1,5	1
Palmitic Acid	1,5	0.4
Stearic Acid	1	0.5
2,2'-Bipyridine	4	4

[a] 1 - Lake Marion, 2 - Fort Loudon Lake, 3 - Holston River, 4 - Mississippi River, 5 - Watts Bar Lake

TABLE 67. PCB CONCENTRATIONS IN SELECTED WATER COURSES (146)

State	Concentration Water µg/ℓ	Concentration Sediment µg/kg
Alaska	ND	ND
Arkansas	ND	20-2,400
California	0.1, 0.1	20-190
Connecticut	0.1-0.2	40
Hawaii	ND	ND
Georgia	--	10-1,300
Maryland	0.1	10-1,200
Mississippi	ND	50; 170
New Jersey	0.1	8-250
Oregon	ND	15; 140
Pennsylvania	0.2	10-50
South Carolina	--	30-200
Texas	0.1-3.0	7.9-290
Virginia	0.1	5-80
Washington	ND	ND
West Virginia	ND	10

PCB's are thus quite persistent in water/sediment systems, and lifetimes of years or even decades have been postulated (481).

The continued presence of PCB's makes it inevitable that they will enter the food chain. As they tend to accumulate in lipid tissues in higher plants and animals, it has been estimated that PCB's will concentrate up the food chain to as much as 10^7 times the water concentration (482).

BIOLOGICAL CONTAMINANTS

An important pathway for certain communicable disease transmission to man is the consumption of contaminated water. Direct disposal of wastewater is the principal contamination route. Land disposal of wastewater is not an important pathway, as pathogenic organisms have limited mobility in soil and seldom migrate far enough to contaminate water supplies (296). However, in contrast to their restricted mobility in soils, biological contaminants are readily dispersed and transported by receiving waters. Consequently, there is a high potential for direct public contact through drinking or recreational use. Wastewater treatment has diminished this threat by reducing the number of organisms in the wastewater. This, combined with natural pathogen mortalities, has greatly lessened the outbreak of water-borne diseases attributable to public water supplies.

The environmental factors that influence the survival of pathogens in fresh water are, in most cases, similar to those that prevail in marine systems. These factors will be discussed in greater detail in the marine water section of this chapter. Briefly, however, the chief factors influencing survivability are temperature, pH, sunlight, toxins, predators, and lack of nutrition, which affect pathogens to different degrees. An examination of Table 68 reveals that pathogens may survive for long periods of time and travel great distances before destruction by environmental factors.

Pathogenic bacteria are best adapted to survival in the human body or to conditions resembling those found in the body. Consequently, natural water systems are a hostile environment. However, cool water is generally more hospitable than warm water because of the depressed metabolism of both the bacteria and their predators. Predatory organisms, especially in slightly polluted waters, are a major contributor to bacterial die-off. For instance, Barua (35) noted that *Vibrio cholerae* survived one to two weeks in clean water as opposed to one to two days in water with a large bacterial population.

Pathogenic bacteria also suffer from a lack of proper nutrition in clean waters; low nutrient levels prevent reproduction. Since die-off rates exceed growth rates, the overall

TABLE 68. AVERAGE TIME IN DAYS FOR 99.9% REDUCTION OF ORIGINAL TITER OF INDICATED MICROORGANISMS IN WATERS (595)

Microorganism	Little Miami River (Ohio)			Ohio River (Ohio)		
	28°C	20°C	4°C	28°C	20°C	4°C
Poliovirus I	17	20	27	11	13	19
ECHO virus 7	12	16	26	5	7	15
ECHO virus 12	5	12	33	3	5	19
Coxsackie virus A9	8	8	10	5	8	20
Aerobacter aerogenes	6	8	15	15	18	44
Escherichia coli	6	7	10	5	5	11
Streptococcus fecalis	6	8	17	18	18	57

population will decline. Other factors affecting die-off are ultraviolet radiation in sunlight, pH extremes, natural antibiotics, and chemical toxins.

In contrast to bacteria, viruses do not multiply in water and, therefore, their number in a water body can never exceed the number introduced into that body by waste disposal. Typically, viruses are much more resistant to external environmental factors (chemical content, pH, temperature, time, etc.) and survive longer than bacteria (136, 198). It was long suspected that algae could inactivate viruses through some process because of low virus concentrations in algae-rich waters. However, it is now believed that the high pH and dissolved oxygen in the vicinity of algal blooms are responsible for the inactivation.

Virus inactivation in lake water is further enhanced by the presence of proteolytic bacteria which degrade the viral coat (292). Coxsackie is particularly susceptible to proteolytic bacteria, while polio virus is generally resistant except to Pseudomonas aeruginosa (292). Otherwise, the mechanisms of virus removal are obscure. Table 69 reports survival times for various enteric viruses in fresh-water bodies.

Transport mechanisms for pathogens include physical current motion, organism motility, adsorption, ingestion, and aerosolization. As most pathogens readily adsorb onto suspended matter, sediment pathogen concentrations may greatly exceed water concentrations. Filter feeding organisms, such as fresh-water shellfish, tend to concentrate pathogenic organisms. Consequently, shellfish can be a major factor in the spreading of certain communicable diseases.

TABLE 69. SURVIVAL OF ENTERIC VIRUSES IN WATER (14)

		Temperature °C		
Type of Water	Virus	4-6	15-16	20-25
		(Range of Days)		
River water	Coxsackie B-3	75/3	8/3	2/3
	ECHO 5	7/0.5	-	3/3
	Polio 1	7/1	-	3/3
	Coxsackie B-3	7/1.7	-	3/3
	ECHO 12	19/3	-	5/3
	ECHO 7	15/3	-	7/3
	Coxsackie A-9	10/3	-	8/3
	Polio 2	75/3	15/3	8/3
	Polio 3	30/3	8/3	8/3
	ECHO 5	60/3	15/3	8/3
	Coxsackie A-9	20/3	-	8/3
	ECHO 12	33/3	-	12/3
	Polio 1	19/3	-	13/3
	Polio 1	60/3	45/3	16/3
	ECHO 7	28/3	-	16/3
	Polio 1	27/3	-	20/3
	Polio 3	50/3	18/3	-
	Polio 3	67/3	7/1.3	7/2.1
	Coxsackie A-2	-	-	5/2
	Coxsackie A-2	-	-	47/2
	Coxsackie B-5	-	24/1	-
	Coxsackie B	18/2	-	-
Impounded fresh water	Coxsackie B-3	7/1.7	-	3/3
	Polio 1	7/1.5	-	3/3
	ECHO 7	22/3	-	4/3
	ECHO 6	5/3	-	5/3
	Coxsackie A-9	6/3	-	6/3
	Polio 1	27/3	-	6/3
	Coxsackie ?	18/3	-	8/3
	ECHO 12	14/3	-	6/3
	Polio ?	21/3	-	10/3
	ECHO ?	23/3	-	12/3
	ECHO ?	21/3	-	20/3
	Polio ?	52/3	-	21/3
	Polio ?	52/3	-	22/3
	ECHO ?	42/3	-	24/3

SECTION 9

CONVENTIONAL WATER TREATMENT: CHEMICAL COAGULATION AND FLOCCULATION FOLLOWED BY SOLIDS SEPARATION

INTRODUCTION

Chemical coagulation and flocculation, followed by clarification or filtration, is common water treatment practice for the treatment of surface waters. The primary purpose is to remove suspended and colloidal solids.

The overall process takes place in three distinct phases. Coagulation involves destabilization of the colloids by rapid mixing of the chemical coagulant with the water in some type of agitated rapid mix tank. Retention time in rapid mixing is very brief, on the order of a few minutes. Flocculation follows in which the wastewater is gently stirred with paddles, allowing the particles to collide and aggregate into larger flocs. Depending on temperature, concentration of the solids, and the type and dosage of coagulant, flocculation requires from 15 minutes to one hour. Clarification and/or filtration usually follows, to provide solids separation.

Commercially available flocculator-clarifier units (often called solids contact units) combine all three operations in a single compartmented tank. In a typical design, coagulant is fed and mixed with the wastewater at the influent pipe; flocculation occurs in a central cone-shaped skirt where a high floc concentration is maintained. Flow passing under the skirt passes up through a solids blanket and out over surface weirs. These units are particularly advantageous for lime softening of hard water, since the precipitated solids help seed the floc, growing larger crystals of precipitate to provide a thicker waste sludge. Recently, flocculator-clarifiers have been receiving wider application in the chemical treatment of industrial wastes and surface water supplies. The major advantages promoting their use are reduced space requirements and less costly installation. However, the unitized nature of construction generally results in a sacrifice of operating flexibility.

The primary substances used as coagulants and their reactions are described below:

1. Aluminum sulfate + calcium bicarbonate
   $Al_2(SO_4)_3 + 3\ Ca(HCO_3)_2 = 3\ CaSO_4 + 2\ Al(OH)_3 + 6\ CO_2$
2. Aluminum sulfate + sodium aluminate:
   $6NaAlO_2 + Al_4(SO_4)_3 \cdot 18\ H_2O + 8Al(OH)_3 + 3Na_2SO_4 + 6H_2O$
3. Aluminum chloride (used under exceptional circumstances only):
   $2\ AlCl_3 + 3\ Ca(HCO_3)_2 = 2\ Al(OH)_3 + 3\ CACl_2 + 6\ CO_2$
4. Aluminum sulfate + hydrated lime:
   $Al_2(SO_4)_3 + 3\ Ca(OH)_2 = 3\ CaSO_4 + 2\ Al(OH)_3$
5. Ferric sulfate:
   $Fe_2(SO_4)_3 + 3\ Ca(HCO_3)_2 = 2\ Fe(OH)_3 + 3\ CaSO_4 + 6\ CO_2$
6. Ferric sulfate + hydrated lime:
   $Fe_2\ (SO_4)_3 + 3\ Ca(OH)_2 = 2\ Fe(OH)_3 + 3\ CaSO_4$
7. Ferrous sulfate:
   $FeSO_4 + Ca(HCO_3)_2 = Fe(OH)_2 + CaSO_4 + 2\ CO_2$
8. Ferrous sulfate + hydrated lime:
   $FeSO_4 + Ca(OH)_2 = Fe(OH)_2 + CaSO_4$
9. Ferrous sulfate + chlorine
   $2\ FeSO_4 + 3\ Ca(HCO_3)_2 + Cl_2 = 2\ Fe(OH)_3 + 2\ CaSO_4 + CaCl_2 + 6\ CO_2$

The most commonly used coagulant is $Al_2(SO_4)_3 \cdot 18\ H_2O$, which is known as filter alum. The amount of hydrolysis which occurs when filter alum is introduced to the water is a function of the pH of the water, with optimum efficiency achieved at a pH of 7 to 8(506).

Because chemical coagulation and clarification is probably the most popular water treatment technique, there exists a substantial volume of information on this technology. Table 70 summarizes the available literature located during the study. As shown in the table, most research work has been performed on the elemental group of contaminants as many of the metals are most efficiently removed by chemical precipitation. Substantial study of turbidity removal and virus inactivation/removal have also been conducted. With the current focus on synthetic/organic and biocidal contaminants in water supplies and their potential carcinogenic effects with long-term ingestion, a surge in research in these areas is anticipated.

TABLE 70. LITERATURE PERTAINING TO CHEMICAL COAGULATION AND CLARIFICATION

Contaminants	Reference Number
Water Quality Parameters	
Asbestos	374, 375
COD	575
Color	226, 414, 575
Hardness	141
Suspended solids	93
Turbidity	9, 137, 157, 225, 226, 274, 548, 575
Elemental Contaminants	
Antimony	537
Arsenic	39, 99, 157, 275, 280, 396, 491, 576, 577
Barium	39, 396, 506, 588
Cadmium	39, 157
Chromium	157
Cobalt	470, 480, 537
Iron	130, 157
Manganese	130, 157
Mercury	157, 394, 506
Molybdenum	537
Nickel	537
Selenium	157, 506
Vanadium	537

TABLE 70 (continued)

Contaminants	Reference Number
Biocidal Contaminants	
DDT	537, 546
Dieldrin	537, 546
Endrin	537, 546
Lindane	546
Parathion	40
Synthetic/Organic Contaminants	
PAH (Polynuclear Aromatic Hydrocarbons)	284
PCB	352
TTHM (total trihalomethanes)	632
Biological Contaminants	
Bacteria	14, 56, 112, 113, 116, 198, 435, 547, 575, 639, 643, 700
Virus	112, 113, 547

WATER QUALITY PARAMETERS

Asbestos has recently been implicated as a carcinogen of potential danger to workers breathing the fiber in asbestos manufacturing plants. Thus, it is feared that the incidence of asbestos in drinking water supplies may also be a health hazard.

Several studies have investigated the ability of chemical coagulation followed by clarification and/or filtration to remove asbestos from water intended for potable purposes. Lawrence et al. (375) examined the effectiveness of various methods: straight filtration, diatomaceous earth filtration, chemical coagulation and combinations. The most effective method involved chemical coagulation with iron salts and polyelectrolytes followed by filtration, which resulted in better than 99.8 percent removal from water containing 12 x 10^6 fibers/ℓ . The optimum ferric chloride dosage was found to range from 6 to 8 mg/ℓ ; satisfactory flocs were formed at all test temperatures. Other reported removal efficiencies were reported as follows:

Treatment	% Asbestos Fiber Removal
. Ferric chloride, polyelectrolyte coagulation and filtration	99.8
. Graded sand filtration only	90
. Diatomaceous earth filtration only	96.8
. Bentanite clay, polyelectrolyte coagulation and filtration	99

As a continuation of the above effort, Lawrence and Zimmermann (374) studied the optimization of alum and polyelectrolyte coagulation for asbestos removal. Optimum removals were obtained with alum concentrations of 30 to 50 mg/ℓ and polyelectrolyte values of 0.3 to 0.6 mg/ℓ . Asbestos removals of over 99.9 percent were achieved. Rapid coagulation and direct filtration was also evaluated and the performance found to be comparable to conventional treatment employing flocculation and sedimentation. In addition, a survey of turbidities and fiber concentrations for several municipal water supplies indicated no correlation between these two parameters.

Shelton and Drewry (575) evaluated the effectiveness of various coagulants in the removal of COD from a raw surface water supply. Results are summarized in Table 71.

TABLE 71 . A COMPARISON OF THE EFFECTIVENESS OF THE COAGULANTS TESTED ON THE RAW SURFACE WATER (575)

Coagulant and Coagulant Aids	Coagulant con- centration mg/ℓ	Maxi- mum Turbidity Removal percent	Coagulant con- centration mg/ℓ	Maxi- mum COD Removal percent	Optimum Dosage* A	B
$Al_2(SO_4)_3$	15	96.50	20	43.0	12	14
$FeCl_3$	34	94.80	48	73.0	37	32
$Fe_2(SO_4)_3 \cdot nH_2O$	35	90.50	38	88.50	65	42
Cationic flocculant	0.56	46.0	0.51	26	†	1.5
$Al_2(SO_4)_3$ + Cationic flocculant	8 + 0.25	97.4	8 + 2.25	49.5	9.2 0.25	7.2 0.25
$Al_2(SO_4)_3$ + Nonionic aid 1	10 + 0.25	97.4	10 + 0.5	59.3	9 0.4	9 0.16
$Al_2(SO_4)_3$ + Anionic aid 1	10 + 0.2	99.65	10 + 0.4	97.5	8.2 0.4	‡ ‡
$Al_2(SO_4)_3$ + Anionic aid 2	9 + 0.2	98.6	10 + 0.2	79.5	7.1 0.2	‡ ‡
$Al_2(SO_4)_3$ + Nonionic aid 2	10 + 0.2	97.20	7 + 0.4	85.00	6.4 0.4	5.4 0.4
$Al_2(SO_4)_3$ + $Na_2OAl_2O_3$	10 + 12	98.70	10 + 9	86.00	10 9	‡ ‡

* Isoelectric point as indicated by: (A) zeta potential, (B) colloidal titration.

† Isoelectric point was not reached with test dosage.

‡ More than one isoelectric point was indicated.

They concluded that aluminum sulfate was the most effective coagulant for COD removal. Cationic polyelectrolyte additions were ineffective while anionic aids gave better results.

Color is caused by humus, tannins, weeds, algae, soluble wastes, and to a certain extent, metals (632). In itself, color is not a health hazard; however, it signals the presence of dissolved organics and metals that may be of some health concern. The above researchers evaluated color removal in the same investigation (575). Ferric chloride was found to perform erratically at differing dosages. Excellent removals were achieved at a 35 mg/ℓ, but at dosages around 35 mg/ℓ and 50 mg/ℓ, color was significantly increased. A study by Mangravite et. al (414) showed that removal of humic acid (one source of color in water supplies) by microflotation produced the same high percentage of removals as the conventional coagulation/sedimentation technique, but at a rate of 5 to 13 times faster. Fulton and Bryant (226) investigated the use of alum with 18 coagulant aids for color removal. Optimum alum dosages ranged from 20 to 25 mg/ℓ and successfully lowered the color concentration to below five standard units. The use of iron salts, oxidation, adsorption, or polymers alone could not achieve this level of removal.

Hardness removal (Ca^{++} and Mg^{++} ions predominantly) via lime-soda precipitative softening is a common practice at many water treatment facilities. Soft water has generally been considered beneficial in that it reduces the usage of soap and detergents, tastes better, and reduces scaling and precipitation in pipes and water heaters. However, some recent studies, summarized by Cooper (141) have discovered an apparent inverse relationship between hardness and heart disease, i.e., people drinking soft water showed a higher incidence of heart disease than those using hard water supplies. In light of these results, the overall benefit of the softening process may have to be reevaluated.

Suspended solids removal from water supplies by coated and uncoated diatomite filter was evaluated by Burns et al. (93). They found that special polyelectrolyte coatings were useful for a final polishing filter process, but that these aids were not as effective as conventional filter aids where large amounts of suspended solids had to be removed.

Turbidity is a uniformly measured water quality parameter inversely related to the visual clarity of the water. Turbidity is caused by clay and other colloidal matter which in themselves are of no health concern. However, heavy metals, virus, and bacteria may be adsorbed onto the clay particles, and the removal of turbidity can be of importance in regards to potential long-term health effects. Because of its popularity as a quality test, many studies or turbidity removal have been performed as summarized below. Robinson (548 compared the effectiveness of alum versus polyelectrolytes for removal of turbidity from muddy surface water. Figure 4 summarizes the performance he reports.

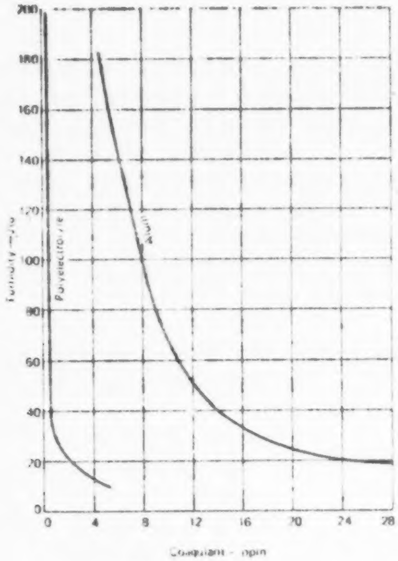


Figure 4. The effectiveness of a small amount of N-607 polymer relative to Alum for raw water with a turbidity of 1-250 Jtu

Fulton and Bryant (226) investigated turbidity removal at a pilot plant treating water from the Croton watershed of New York City. They found that although alum coagulation (with coagulant aids) could produce water below the 1.0 TU Federal Standard, the American Waterworks Association (AWWA) goal of 0.1 TU could not be achieved without further treatment. Al-Layla and Middlebrooks (9) studied the optimum values for operational variables in turbidity removal (alum dosage, flocculation time, and paddle speed).

The following conclusions were drawn from this research:

1. The most significant independent variable was the paddle speed followed by flocculation time and alum dosage.

2. All variables and their first order interactions were significant at the 1 percent level.

3. The optimum alum dosage was 25 mg/ℓ for the conditions under which the study was performed.

4. The optimum paddle speed ranges between 40 and 50 rpm.

5. The optimum flocculation time was approximately 30 min.

6. This study revealed that the significant interactions between the independent variables (alum dosage, flocculation time, and paddle speed) can be utilized in more efficient water treatment. For example, a higher percentage removal of turbidity can be obtained by using low alum dosage and increasing the flocculation time and/or paddle speed. On the other hand, high alum dosages and reduced flocculation times and/or paddle speeds can be utilized.

7. The study shows the importance of the following equipment and methods in the removal of turbidity in water treatment plants:

    a. Variable speed flocculator paddles.

    b. Multiple flocculation basins that can be used in series or parallel operation.

    c. The use of jar tests to determine the optimum alum dosage continuously during the operation of water treatment plants.

Gruninger et al. (274) compared the performance of ferric chloride versus alum as the primary coagulant and found that the combination of 8 mg/ℓ $FeCL_3$ + 4 mg/ℓ bentonite clay + 0.25 mg/ℓ polymer provided comparable performance to the system with alum and clay; there was approximately 95-98 percent turbidity removal.

Reference 157 summarizes the performance of various treatment technologies in removing turbidity. These results are -

- Plain sedimentation: 50 to 95 percent, depending totally on the settling characteristics of solids;
- Coagulation with sedimentation: 80 to 99 percent;
- Rapid sand filtration: 80 to 99 percent with influent turbidity of 10 JTU or less;
- Slow sand filtration: 80 to 99 percent with influent turbidity of 10 JTU or less;
- Diatomite filtration: 80 to 99 percent with influent turbidity of less than 10 JTU.

Shelton and Drewry (575) analyzed the effectiveness of different cationic, nonionic, and anionic polyelectrolytes in reducing turbidity. Table 71, presented earlier, displays their results. Aluminum sulfate with anionic coagulant aids was found to be the most effective, achieving 99+ percent removal at a 10 mg/ℓ dosage. Frissora (225) evaluated the performance of rapid multimedia filtration alone on turbidity removal and achieved removals of 96 to 99.7 percent at high loading rates of 4 to 16 gpm/ft^2. Conley (137) also studied rapid filtration preceded by chemical coagulation and settling, for turbidity removal at a number of water treatment plants around the country. Table 72 summarizes the results of his study; removals ranged from 80 to 97 percent and averaged 94 percent.

ELEMENTAL CONTAMINANTS

Many elemental contaminants are readily removed by chemical coagulation and subsequent solids separation steps (settling or filtration). As some of the elemental contaminants are being more closely reviewed for possible long-term health effects, the importance of chemical coagulation as a removal process becomes more pronounced.

TABLE 72. TYPICAL THREE-MEDIA HIGH-RATE FILTRATION PLANT PERFORMANCE

	Turbidity				
Location	Applied JTU	Filtered JTU	Filter Rate gpm/ft^2*	Filter Runs hr	Backwash Water percent
Regina, Sask.	0.5	0.1	4	30	2
Siler City, N.C.	1	0.2	5	30	2
Pasadena, Tex. (industrial)	3	0.5	6	40	1
Corvallis, Ore.	3	0.2	3	50	1
Ocaquan, Va.	1	0.1	4	48	1
Samoa, Ca. (industrial)	10	0.5	5	18	2
Fort Lauderdale, Fla.	1	0.1	4	80	1
Knoxville, Tenn.	1	0.2	5†	60	1
Albany, Ore.	2	0.2	5	20	2
Longview, Tex.	4	0.2	3	48	2
Paris, Tex.	10	0.3	3	50	2
Newport, Ore.	3	0.2	4	20	3
Norristown, Pa.	6	0.3	4	45	1
Springfield, Mo.	1.5	0.2	4½	24	1
Winnetka, Ill.	2	0.3	5	15	2
Stanton, Del.	2	0.2	4	30	3

* Typical summer peak daily flow rate. Hourly peaks are generally in the range of 4-6 gmp/sq ft.

† Under test. Some filters are out of service so that rate could be increased on remaining filters.

Arsenic is one element being closely watched in water supplies as it has a relatively high toxicity, accumulates in the body, and has been associated with the occurrence of cancer (39). Several studies on arsenic removal are available. Some 1,056 cases of black-foot disease and skin cancer have been reported in the southwest part of Taiwan. According to statistical data, there is a close relationship between these diseases and the high arsenic content (0.6 to 2.0 mg/ℓ) in deep-well water used for drinking (577). Since there is no other available water source in the area, some practical and economical method to remove the arsenic compounds was urgently needed. To satisfy this need, Shen (576) performed a lengthy analysis of treatability tests to evaluate the arsenic removal capability of coagulation/settling/filtration processes. Initial coagulant tests showed ferric chloride to be the best chemical, achieving 92 percent As removal at a 30 mg/ℓ dosage. Subsequent testing, however, showed that these removals could be improved by preoxidation before coagulation. Adding 20 mg/ℓ of chlorine and then coagulating with 50 mg/ℓ of ferric chloride provided the best results, achieving 98.7 percent As removal.

Gulledge and O'Connor (275) showed that As removal was affected by pH and doses of suitable coagulants. Arsenic adsorption onto ferric hydroxide exceeded adsorption onto aluminum hydroxide. For both coagulants, increased dosages (up to 50 mg/ℓ) resulted in increased removal. A significant decrease in arsenic removal was seen at pH over 8.0.

Logsdon et al. (396) studied arsenate removal by precipitation with ferric sulfate and alum, in pilot plant tests. For initial arsenic concentrations of 0.3 mg/ℓ, ferric sulfate treatment yielded 91 to 94 percent removal. Dual media filtration achieved an additional 5 to 7 percent removal for overall removals of 98 to 99 percent and effluent levels of 0.003 to 0.006 mg/ℓ arsenic. Alum treatment was less effective, yielding 75 to 79 percent removal without filtration and 85 to 92 percent removal with filtration (280). Arsenite removals of only 10 to 25 percent were achieved by alum, and 40 to 60 percent by ferric chloride. However, when chlorination preceded coagulant addition, removals equivalent to those reported for arsenate were observed (280). Chlorine apparently oxidized the arsenite to arsenate. For ferric sulfate precipitation, treatment efficiency remained constant from pH 5.5 to 8.5. Above this pH, efficiency declined. With alum, optimal precipitation was at pH 7.0. with slight reduction in efficiency at pH below 7.0, and rapid drop-off in removal efficiency at pH above 7.0. Similar pH effects for arsenate treatment by iron and aluminum salts have been observed in another study (491). Calmon (99) found that cold lime-soda precipitation with filtration was capable of removing 96.4 percent of the arsenic, whereas ferric sulfate removed less than 50 percent.

The toxicity of antimony is similar to arsenic, although less acute. Very little research has been conducted related to Sb, as other metals appear to be of more importance from a health-effects standpoint. Lime coagulation has been shown to provide Sb removals of better than 90 percent from wastewater effluent (537). Similar removals may be possible in water supply treatment, but no data was found.

Barium is a toxin that acts as a muscle stimulant. As such, it strongly affects the heart muscles and causes high blood pressure and nerve blockage (39). Barium removal - only for wastewater treatment - was reported by several researchers. Sigworth and Smith (588) found during pilot wastewater plant tests that lime coagulation and settling was an effective method, resulting in a 99 percent Ba removal. Dosage rates were 45 mg/ℓ $FeCl_3$, 260 mg/ℓ lime plus 20 mg/ℓ $FeCl_3$ for the low lime test, and 600 mg/ℓ lime for the high lime run. Logsdon (396) found improved removals with the lime softening process. Treatment efficiency was pH dependent, increasing to a maximum of 98 percent at pH 10.5, and dropping off at higher pH values. Barium is not appreciably removed by ferric or alum coagulation.

Cadmium has a high toxic potential, accumulates in the kidney and liver, and has been suggested as a causative factor in high blood pressure (157). Only minute traces of cadmium are found in natural waters. However, several industries discharge cadmium in their wastewaters, including metallurgical alloying, ceramics manufacture, electroplating, inorganic pigments, textile printing, chemical industries, and acid mine drainage. Very little data are available regarding the use of coagulation for Cd removal at water treatment plants. The summary reference (157) lists removals at 50 to 90 percent with coagulation followed by filtration. Unpublished data developed by EPA indicates that lime softening and ferric sulphate coagulation above pH 8 achieves Cd removals of over 98 percent.

Chromium in the hexavalent form is highly toxic to man and has been shown to be carcinogenic to man when inhaled. Volkert and Associates, in a study for EPA (157), summarized that coagulation and sedimentation with filtration is capable of removing 50 to 90 percent of the insoluble chromium (Cr) forms present in water supplies.

The hexavalent form is much more difficult to remove with typical water treatment coagulants than is the trivalent form. It is desirable, therefore, to reduce the chromium present to trivalent prior to treatment. Laboratory studies by EPA (unpublished) indicate that trivalent chromium is removed at better than 95 percent levels by ferric sulphate up to pH 9.5, removed at 90 percent levels by alum up to pH 8.5, and removed at 98 percent level by lime softening at pH from 10 to 11.5. Hexavalent chromium, however, is not effectively removed by any of these coagulants. Cobalt (Co) concentration in potable water sources seldom reaches levels that require special consideration. Cobalt is currently best treated by lime coagulation, although as much as 2 mg/ℓ may be left in the effluent (537). Nilsson (480) found that lime coagulation was capable of achieving 88 percent removal of cobalt. Another study (470) found that cobalt precipitation could be enhanced by the addition of chitosan or a chelate as a polishing agent. An EPA study (537) concluded that similar precipitation techniques are also applicable to molybdenum (Mo), nickel (Ni), and vanadium (V) removal.

Iron and manganese, often found together, are important constituents of potable water supplies because they can import unwanted aesthetic qualities. From a health standpoint, however, iron and manganese do not pose any significant threat in the concentrations normally found in surface and groundwater supplies. Therefore, removal of these constituents will not be discussed in any depth here. Effective removal can be provided by aeration or more commonly, by chemical precipitation, sedimentation, and filtration (130).

Recent attention has been focused on the contamination of water supplies by organic and inorganic mercury (Hg) by industrial discharges. Fortunately, recent rests by the EPA of 273 water supplies across the country showed very low mercury levels in nearly all of them. Even so, the performance of conventional water treatment technologies and new techniques for removing Hg from water is of interest. Logsdon and Symons (394) investigated the efficiency of conventional water treatment processes in removing Hg. Bench scale studies of chemical coagulation and settling yielded the following conclusions:

1. Mercury removal during coagulation was related mainly to adsorption of mercury onto the turbid-causing agents in the water.

2. In raw waters with low turbidity, ferric sulfate was more effective than alum for removal of inorganic mercury. Removals ranged from 40 to 60 percent over a turbidity range of 2 to 100 JTU's.

3. Removal by coagulation or adsorption onto turbidity-causing agents was less for methyl mercury than for inorganic mercury.

4. Inorganic mercury removal by softening was most effective in the pH range of 10.7 to 11.4 and is thought to be related to adsorption onto magnesium hydroxide floc.

5. Methyl mercury was not removed by softening.

The authors concluded by stating that as long as environmental levels of mercury in raw water remain low (near drinking water standards), extremely high removals will not be required, and conventional technology should be sufficient.

Molybdenum can occur naturally in drinking water supplies at concentrations up to 20 μg/ℓ due to weathering of minerals. It can also be found in water as a result of waste discharges from industries related to the manufacture of glassware, ceramics, printing inks, electrical equipment, and certain steel alloys. Virtually no data relative to molybdenum removal from water supplies are available, because molybdenum is not currently seen as a primary health hazard.

Nickel occurs naturally at concentrations up to 0.072 mg/ℓ with an average concentration of about 0.005 mg/ℓ. Wastewater discharges from industries associated with nickel-plating, nickel alloying, storage battery manufacturing, organic hydrogenation, and the manufacture of nickel-chrome resistance wire can contribute to the presence of nickel in water sources (21). In large doses, Ni can be harmful; however, in this country, natural concentrations are low, and lime coagulation appears to be successful in removing large percentages of Ni from wastewater. No literature pertinent to Ni removal by water treatment facilities was found.

Alum and ferric sulfate coagulation and lime softening are only moderately effective for selenite (Se^{+4}) removal, and are largely ineffective for selenate (Se^{+6}) removal (396). Studies with ferric sulfate (30 mg/ℓ) at a pH of 5.5 yielded removals of 85 percent from river water containing 0.03 mgSe^{+4}/ℓ. Removals decreased as the pH increased. The maximum achievable Se^{+4} removal with alum (100 mg/ℓ, pH 6.9) was 32 percent. A maximum removal of 45 percent was obtained with lime softening. No conventional method achieved greater than 10 percent removal of Se^{+6}.

No data were located relative to vanadium removal from water supplies. In wastewater treatment, however, very good vanadium removals were achieved via coagulation with iron or aluminum salts or chitosan polymers (537).

BIOCIDAL CONTAMINANTS

Recently, there has been increased awareness of the potential health hazards of biocidals. Limited data, however, are available in the literature regarding the removal of these constituents from water supplies by chemical coagulation: Robeck et al. (546) studied the effects of various water treatment processes on pesticides. Their results, in regards to pesticide removal via chemical coagulation (alum) and filtration, are shown in Table 73 below.

TABLE 73. PERCENTAGE OF PESTICIDE REMOVED BY CONVENTIONAL WATER TREATMENT (546)

Pesticide	Load - ppb			
	1	5	10	25
Lindane	<10	<10	<10	
Dieldrin	55	55	55	
DDT			98	97
Parathion			20	
2, 4, 5-Tester			63	
Endrin	35		35	

* Total hardness as $CaCO_3$ reduced from 260 to 33 ppm and pH increased from 7.6 to 10.4.

As can be seen, DDT was readily removed, whereas lindane, parathion, and endrin were not. Dieldrin and 2, 4, 5-Tester are removed at slightly better than 50 percent. Softening with lime and soda ash and with an iron salt as a coagulant did not improve on the removals obtained with alum coagulation alone.

Another study (380) evaluated the effect of $KMnO_4$ as an oxidant and precipitating agent. It was found that $KMnO_4$ was not significantly effective in removing lindane, but was capable of removing over 80 percent of the heptachlor present in under 5 hours. Removals of DDT were less than 20 percent in 48 hours, and endrin was totally unaffected by the process.

SYNTHETIC/ORGANIC CONTAMINANTS

In recent years, concern has been expressed over the possible occurrence of certain carcinogenic compounds in drinking water (World Health Organization, 1964). A group of compounds which has received particular attention is the polynuclear (polycyclic) aromatic hydrocarbons (PAH), some of which are potent carcinogens under certain conditions. It is, however, far from certain that these compounds are significant when present in the trace amounts found in drinking water. Clearly, further research is needed into both the levels and health effects of PAH in the environment (284).

Harrison et al. conclude (284): 'The reliability of much of the information concerning the removal of PAH by conventional water treatment processes is open to considerable doubt. Field experiments have frequently ignored retention times within the works, and hence been rendered unreliable at the sampling stage. Laboratory studies have commonly used unrealistic high concentrations of PAH, and the high removals achieved may be largely explained often by the low solubility of the PAH themselves. Further analytical work is required in this field, and fundamental studies of the chemical changes that occur upon chlorination of these compounds at low concentrations are necessary. Increasing water reuse makes the need for this type of information particularly acute."

After the EPA disclosed its findings of organic chemicals in New Orleans' water, it was decided to conduct another, more inclusive study at specific sites across the U.S. In this study, Symons et al. (632) found that precipitative softening at water treatment plants increased the concentration of trihalomethane in the product water from an average of 0.49 um/ℓ (80 locations) to 0.84 um/ℓ (17 plants with softening). This indicates that chlorination at a higher pH will produce higher concentrations of trihalomethanes.

Kinoshita and Sunada (352) performed an experiment on the removal of PCB by chemical coagulation. Bentonite, sodium carbonates, and aluminum sulfate were added to a solution of 100 ppb PCB. After settling, the PCB concentration has been reduced 90 percent, suggesting that conventional water treatment by coagulation and settling or filtration provides a margin of safety against the ingestion of PCB's in raw water supplies.

BIOLOGICAL CONTAMINANTS

The use of diatomite filtration with and without chemical addition is capable of removing bacteria from water supplies. McIndoe (435) summarized research in this area as follows:

- Work by the U.S. Army Engineering Research and Development Laboratories at Fort Belvoir, Virginia, showed that a diatomite filter using a coarse grade of filter aid was capable not only of 100 percent removal of the entamoeba cysts, but also of partial removal of other pathogenic organisms. Work done at Rutgers University has shown that even the coarsest grade of diatomite gave coliform removals of greater than 50 percent for influent levels of 210 to 1300/100 mℓ. Finer grades gave "complete" coliform removals of influent levels up to several thousand per 100 mℓ.

- Work by Chang (112, 113), Hunter (313), and others with solutions of iron salts added to bacteria-contaminated water has indicated a reaction takes place between the iron ions and the protein sheath of both bacterial and viral organisms. Subsequent removal of the iron by the diatomite MgO process has produced total count reductions on the order of 99 percent, with fecal strep and Escherichia coli counts of zero.

- Waters so clarified would be expected to be readily disinfected by usual practices.

CONTAMINANTS

Amirhor and Englebrecht (14) analyzed the potential use of uncoated and polyelectrolyte -aided diatomaceous earth filtration for bacterial virus removal. They concluded that uncoated DE filtration was not effective for virus removal, but that precoating of cationic polyelectrolyte greatly improved removals. Certain variables such as pH, level and concentration of polyelectrolyte coating, and virus concentration affected removals.

Wolf et al. (700) conducted a large-scale pilot study of virus removal by both lime and alum. They demonstrated that virus removals from secondary effluents by alum coagulation-sedimentation and coagulation-sedimentation-filtration processes are essentially the same as described in the literature using smaller-scale processes. Removals of bacterial virus as high as 99.845 percent for coagulation-sedimentation and 99.985 percent for coagulation-sedimentation-filtration processes were observed at an Al:P ratio of 7:1.

At a lower alum dose there was a marked decrease in virus removals. At an Al:P ratio of 0.44:1, removals of only 46 percent of f_2 coliphage and 63 percent of polio virus by the coagulation-sedimentation process per se were observed.

High lime treatment of secondary effluents achieved very high degrees of virus removal, but the percentage has not yet been quantified.

Englebrecht and Chaudhuri (116) conducted a study to extend the knowledge in this field. Their conclusions follow:

1. Chemical coagulation and flocculation is an effective process in removing viruses from water. Removals in the range of 98.0 to 99.9 percent can be expected.
2. The presence of bivalent cations like calcium and magnesium up to a concentration of 50 mg/ℓ each does not interfere with the efficiency of the process.
3. The efficiency of virus removal is reduced when the raw water contains organic matter.
4. Intelligent use of commercially available cationic polyelectrolytes with or without hydrolyzed metal ions may markedly increase the efficiency of the coagulation and flocculation process in removing virus.
5. Virus particles remain active in the settled sludge following their removal from water by coagulation and flocculation, and can be recovered from the floc by various eluants. Therefore, proper care should be taken in sludge disposal.

Thorup et al. (643) conducted a study to determine the effectiveness of polyelectrolytes used as coagulant aids for the removal of virus from artificially seeded water. They found that the cationic polyelectrolyte performed more acceptably than the anionic and nonionic types and was beneficial in aiding floc formation under conditions of poor coagulation. However, the 80 to 94 percent removal achieved under these circumstances was well below the 99+ percent usually considered acceptable. In instances of adequate coagulation, however, cationic polyelectrolytes did not increase virus removal beyond the levels obtained with unaided $Al_2(SO_4)_3$ or $Fe_2(SO_4)_3$ coagulation.

Shelton and Drewry (575) performed a literature search of virus (f2 bacteriophage) removal via coagulation and, based on the results, studied the effectiveness of different chemical coagulants and polyelectrolytes for this purpose. Their conclusions were:

1. Aluminum sulfate, ferric chloride, and ferric sulfate are the most promising of the group of primary coagulants tested, from the standpoint of f2 bacteriophage removal, all producing greater than 99 percent removal. Aluminum sulfate is considered to be the most effective coagulant, whereas ferric chloride and ferric sulfate are less effective.

2. The cationic polyelectrolyte tested is not satisfactory as a primary coagulant because of its poor floc formation and settling characteristics. Virus removals with the cationic flocculant were only moderate (92 percent).

3. Anionic, nonionic, and cationic polyelectrolytes have only minor significance in virus removal. It is noted, however, that the anionic coagulant aids are useful in widening the effective dosage range for good virus reduction. The nonionic coagulant aids give moderately better results, which are attributable to an ability to form more dense and "sticky" floc. This improvement of virus removal is indirect, because the nonionic polyelectrolytes do not form a virus-ion complex with the virus protein coat. The cationic coagulant aid produces a moderate improvement in virus removal; however, it is questionable if this improvement would be economically justifiable for a full-scale operation.

4. The use of sodium aluminate with aluminum sulfate does cause a marked increase in virus removal, to 99.9 percent overall. This increase, however, could be used only for "special-case" situations, since the sodium aluminate dosage for optimum virus removal is not compatible (by a factor of 3) with turbidity and COD optimums.

Thayer and Sproul (639) of the University of Maine did extensive research concerning the effects of water softening upon virus inactivation. Their primary objective was the determination of the effect of chemical precipitation in water softening upon bacterial viruses. Inactivation of T2 virus varied widely, with the best results, on the order of 99.999 percent inactivation, occurring when magnesium hydroxide was the only precipitate formed. The standard excess lime-soda-ash process also produced good results with about 99.45 percent virus reduction.

Lime flocculation with rapid sand filtration, a long-used standard water-treatment method, was investigated for its virus-removal characteristics by Berg et al. (56) at the Cincinnati Water Research Lab. Conclusions based on their study using polio virus I (nacuine) exclusively were that (1) flocculation of secondary effluent yielded up to 99.86 percent removal of the virus, dependent upon the coagulant dose and the pH level attained, and (2) the total removal by lime flocculation and rapid sand filtration through an 8-in-deep filter achieved a maximum of 99.997 percent.

The Journal of the Sanitary Engineering Division, ASCE, published an article in 1961 summarizing the virus work done up to that time (198). Table 74 shows the results of this work. As shown, the dosage of flocculant alone is not a measure of the efficiency of the process.

Robeck et al. (547) investigated the fate of human viruses in rapid filtration processes with and without flocculation and settling. They found that if low alum doses were fed just ahead of the dual-media filter (operating at 6 or 2 gpm/ft^2), more than 98 percent of the virus was removed. When the alum dose was increased and conventional flocculators and settling added, removals were increased to over 99 percent. When turbid water was treated, a floc breakthrough was usually accompanied by an increase in virus penetration. Polyelectrolyte doses as low as 0.05 mg/ℓ increased floc strength and helped to prevent such breakthroughs.

The current state of knowledge indicates that chemical flocculation, settling, and filtration are effective in removing virus from water. Removals of 99$^+$ percent have been reported under proper operating conditions. However, more research is still needed in the area to fully determine the most effective doses of coagulants and coagulant aids, the physiochemical effects of turbidity, pH, temperature, and colloidal charge, and to develop optimal operating parameters.

Chang et al. (113) performed comprehensive studies of the dynamics of removal of bacterial virus by aluminum sulfate flocculation. From their observations they concluded:

1. Flocculation by aluminum sulfate can remove high percentages of virus, and within the zone of flocculation, higher doses produced greater efficiency.

2. The virus is concentrated in the floc sediment and is not destroyed, but only temporarily inactivated. It will become active again, if dissociated from the aluminum.

TABLE 74. REDUCTION OF HUMAN ENTERIC VIRUSES IN WATER BY CHEMICAL FLOCCULATION (198)

Virus	Type of Water	Stages	Coagulant Added[a] ppm	Floc Amount or Description	Virus Removal
Polio	Tap	1	100		Little[b]
	Raw	1	100		Some[c]
Polio	River	1	136		Some to significant[d]
		1	410		Some
		1	136-273	0.4-1.03 mi per l	Little to some
		1	273-546	1.5-2.2 mi per l	Significant
Infectious hepatitis[e]	Distilled	1	69		Little to some
Coxsackie A5	Spring	1	28-45		Some
Coxsackie A2 (purified)	Ohio River (16-255ppm turbidity)	1	15	Good	95.7% at 25°C
		1	25	Very good	95.9% at 5°C
		2	15	Very good	98.6% at 25°C
			15*	Very good	99.8% at 25°C
		2	25	Very good	99.6% at 5°C
			25*	Very good	99.9% at 25°C

[a] Alum except where marked with an asterisk; asterisks indicate ferric chloride.

[b] Probably less than 25%.

[c] Probably less than 50%.

[d] More than 50%.

[e] Gauze-strained fecal suspension in distilled water.

3. Virus inactivation is believed to be the result of the formation of an aluminum protein salt in the virus.

Chang et al. performed another study (112) evaluating the efficiencies of alum and ferric chloride in removing coxsackie and bacterial viruses. They analyzed the effects of coagulant dosage, pH, and rate and method of stirring on virus removal. A 40 mg/ℓ dose of $Al_2(SO_4)_3$ yielded an 86.3 percent removal of coxsackie virus and a 93.5 percent removal of bacterial virus. Under similar testing conditions, 20 and 40 mg/ℓ of $FeCl_3$ facilitated the following removal percentages: 96.6_4 coxsackie virus and 99.3 bacterial virus, and 98.1_4 coxsackie and 99.9 bacterial virus.

CONVENTIONAL WATER TREATMENT: DISINFECTION

INTRODUCTION

Disinfection refers to the inactivation or destruction of pathogenic microorganisms. Disinfectants (chlorine, ozone, ultraviolet and ionizing radiation) also have secondary applications, particularly as oxidants for the removal of organic contaminants. Both applications are included in the literature pertaining to the treatment of drinking water with disinfectants (Table 75).

In the United States, the traditional disinfectant is chlorine, which is usually applied to the water in gaseous form as the last treatment step. The chlorine gas (Cl_2) reacts with the water to form hypochlorous acid (HOCl), which dissociates at pH values above 7 to form the hypochlorite ion (OCl^-)(332). Occasionally hypochlorous acid, hypochlorite, or chlorine dioxide (ClO_2) are used instead of chlorine gas.

Next to chlorine, the most popular disinfectant is ozone, especially in Europe. Ozone (O_3) is an unstable gas and must be generated at the site and used immediately. It is a powerful oxidant. Both its germicidal and oxidizing properties seem to be the result of the formation of several free radicals in water (HO_2, OH, HO_3^-), which will attack almost all organic compounds.

Ultraviolet and ionizing radiation have been used in pilot plant and small industrial applications. Like ozone, they seem to act by forming a series of free radicals in water, which can attack organic bonds. Ultraviolet is incapable of acting at more than a few centimeters depth, and both forms of radiation are highly susceptible to interference from turbidity and suspended matter. Ionizing radiation requires radioisotopes and the concommitant shielding and elaborate safety precautions.

All of the disinfectants have disadvantages that prevent any of them from being universally applicable. For a given situation the choice depends largely on the water quality, types of microorganisms in the water, desirability of nondisinfection applications, and cost.

TABLE 75. LITERATURE REVIEWED PERTAINING TO WATER DISINFECTION

Contaminant	Reference Number		
	Chlorination	Ozonation	Radiochemical
Water Quality Parameters			
Ammonia	332, 366, 616, 688	332	469
BOD, COD, TOC	332, 555, 616	332, 351, 465, 660	
Nitrites		332	
Elemental Contaminants			
Iron		157, 402, 465	
Manganese		157, 351, 465	
Biocidal Contaminants			
Chlorinated hydrocarbons	91, 157	91, 157, 332	157
Organo-phosphorus			157
Carbamates			157
Synthetic/Organic Contaminants			
PCBs			157, 352
General	284, 319, 555, 616	157, 284, 465	157
Biological Contaminants			
Viruses, general	55, 111, 136, 242, 284, 332, 399, 463, 547, 600, 616	111, 242, 332, 402, 463, 465, 600, 660	
Coliforms	111, 366, 537, 555	61, 111, 283, 332, 537	
E. Coli	55, 332, 351, 660	351, 465, 660	111, 469, 656
Clostridium		660	
Protozoa	111, 660	111, 332, 660	

WATER QUALITY PARAMETERS

Although disinfectants are seldom specifically employed to remove any of the water quality parameters, there are certain chemical reactions that make the idea feasible. The reactions of chlorine with ammonia are the best known. If allowed to go to completion, the ammonia will be oxidized to nitrogen gas and will thus be removed from the water. Ozone does not react with ammonia (332). Ammonia is seldom an issue, though, as it is not a common constituent of most water supplies.

The organics, as represented by BOD, COD, and total organic carbon, in drinking water are susceptible to oxidation by disinfectants. However, reactions other than oxidation may produce potentially hazardous compounds. For instance, Rook (555) and McClanahan (332) reported that chlorine reacted with humic and fulvic substances, forming chlorinated organic compounds. These chlorinated compounds are much more resistant than the precursor compounds to both biodegradation and chemical oxidation. Consequently they persist in a water supply that is not treated any further than chlorination. Some of the chlorinated compounds formed are suspected to have carcinogenic properties.

Ozone is even more reactive toward organic compounds than is chlorine. With ozone, though, the reactions are almost exclusively oxidation, with few if any hazardous compounds formed in side reactions. The formation of ozonides and similar compounds has been postulated, but there has been no evidence to date demonstrating their formation during the ozonation of water. Morris (465) reported an increase in BOD after ozonation and attributed it to the breakdown of nonbiodegradable organic molecules into simple, degradable compounds.

Murphy (469) recently demonstrated that gamma radiation had an oxidizing effect on organic compounds similar to that of ozone. Ultraviolet radiation theoretically has a similar effect.

It should be noted that nonbiological contaminants interfere with the primary disinfection role of these chemicals by consuming the disinfectants. To achieve proper disinfection in highly organic waters, for instance, requires large increases in applied dosages. Some water-borne disease outbreaks are attributed to improper disinfection of highly organic water supplies.

ELEMENTAL CONTAMINANTS

In general, the disinfectants have no effect on elemental concentrations. Elements would have to be in a reduced state before oxidizing disinfectants could have an impact. This is not likely in most drinking water supplies. Exceptions are iron and manganese, which are more soluble in their lower oxidation states. Morris (465) reported that ozone readily oxidized soluble iron and manganese to the insoluble oxides, which could then be removed by sedimentation or filtration.

Trivalent chromium can be oxidized to the hexavalent form, which is more toxic and difficult to remove with conventional coagulation/filtration processes.

BIOCIDAL CONTAMINANTS

Chlorinated hydrocarbon biocides are generally resistant to chemical oxidations. Stone et al. (537) reported that chlorine was not a particularly effective oxidant for such biocides. Ozone was more effective, but removal efficiencies varied widely from 16 to 93 percent, depending on the type of biocide, ozone concentration, and contact time (Table 76). Stone et al. reported other ozone studies that yielded 50 percent removal of endrin, 75 percent removal of lindane, and approximately 100 percent removal of dieldrin and aldrin. They also stated that ultraviolet radiation could completely eliminate carbamate biocides, reduce aldrin by 45 percent, and reduce endrin and dieldrin by 18 percent.

Buescher et al. (91) studied the chemical oxidation of chlorinated hydrocarbons in water. They concluded the following:

- Lindane concentrations in aqueous solutions were readily decreased by ozonation and only partially affected by potassium permanganate. Treatment with chlorination, peroxides, and aeration had no measurable effects.

- Aldrin in aqueous solutions was readily attacked by chlorination, potassium permanganate, ozone and aeration; peroxides had no measurable effects.

- Dieldrin concentrations in aqueous solutions were decreased by ozonation and aeration.

- Lindane and aldrin in aqueous solution were found to be volatile. The degree of volatility may be an indication of the susceptability of that pesticide to chemical treatment.

- Lindane in the presence of other naturally occurring trace organics found in river water was readily attacked by ozonation, though aeration had only a minor effect.

TABLE 76. EFFECT OF OZONATION ON CHLORINATED HYDROCARBON INSECTICIDES (537)

Time of Ozonation	Ozone Absorbed mg/ℓ	γ-BHC		Dieldrin		DDT		TDE (DDD)	
		Before Ozonation	After Ozonation	Before Ozonation	After Ozonation	Before Ozonation	After Ozonation	Before Ozonation	After Ozonation
		µg/ℓ							
5	8.8	1.32	0.88	1.29	1.08				
10	18.3	1.39	0.81	1.30	0.66				
20	36.0	1.31	0.34	1.31	0.22				
5	11.7					2.00	0.54	2.00	0.62
10	20.0					2.00	0.46	2.00	0.43
20	38.2					2.00	0.14	2.00	0.13

SYNTHETIC/ORGANIC CONTAMINANTS

As previously mentioned under "Water Quality Parameters," both chlorine and ozone readily react with dissolved organics. However, synthetic organics are often more resistant to oxidation than the natural organics. Rosenblatt (332) indicated that chlorine reacted with many organics to give both chlorinated and oxidation products, but that there was no reaction with many others (Table 77). Ozone is an effective oxidant against the phenolics and organic nitrogen compounds, but not against many of the simpler organic molecules, such as ethanol. Harrison et al. (284) reported that chlorine was more effective than ozone against benzo(a)pyrene.

Many of these synthetic organics, such as nitrobenzene, benzo(a)pyrene oniline, and ethyl benzene are reportedly carcinogenic. While these chemicals are seldom found in drinking water supplies at concentrations exceeding a few ppm, the postulated no-threshold-dose character of many carcinogens makes even one molecule a potential hazard. Note also that chlorine is suspected of producing chlorinated organic compounds which may themselves be carcinogenic.

Kinoshita and Sunada (352) investigated the effects of irradation on PCB in water. They concluded that PCB in aqueous microparticulate colloidal solution is destroyed by ionizing irradiation (up to 95 percent), but that its resistance to radiation is far greater than other chlorinated hydrocarbons used as pentachlorophenol or DDT, and other pesticides such as parathion. They also found that the acute toxicity of the irradiated PCB solution was far less than the nonirradiated solution for striped shrimps.

BIOLOGICAL CONTAMINANTS

The major application of disinfectants is against biological contaminants. In this light, the disinfectants have been evaluated primarily on their effectiveness in controlling biologicals (e.g., bacteria, viruses, protozoa, parasitic worms).

Chlorine is the traditional disinfectant in the United States. It is effective to some extent against all types of pathogenic organisms found in water. Bacterial kills of at least 99 percent are considered normal (537), and 4 to 5 log reductions are not unusual. Both Sobsey (600) and Long (399) reported virus reductions of up to 99.99 percent. Reference 136 summarized research on virus destruction by chlorine as shown in Table 78. Chlorine can be effective when used with filtration against free-swimming protozoa and parasitic worms. However, chlorine is relatively ineffectual against their ova and cysts that are resistant to oxidation. Chlorine has the further advantage of persistence given a

TABLE 77. PROBABLE REACTION PRODUCTS OF CHLORINE AND SOME TYPICAL ORGANIC COMPOUNDS FOUND IN POLLUTED WATER SUPPLIES (332)

Organic Compound	Probable Reaction Products
Alcohols	
Methanol	None
Isopropanol	None
tert-Batanol	None
Ketones	
Acetone	None
Benzene and Derivatives	
Benzene	None
Toluene	None
Ethylbenzene	None
Benzoic acid	None
Phenol and Phenolics	
Phenol	Mono-, di-, and trichlorophenols; non-aromatic oxidation products
m-Cresol	Mono-, di-, and trichlorocresols; non-aromatic oxidation products
Hydroquinone	p-Benzoquinone, non-aromatic oxidation products
Organic Nitrogen Compounds	
Aniline	Mono-, di-, and trichloroanilines; non-aromatic oxidation products
Dimethylamine	N-Chlorodimethylamine, oxidation products
Nitrobenzene	None

TABLE 78. VIRICIDAL EFFICIENCY OF FREE CHLORINE IN WATER (136)

Investigator	Virus	Temp. °C	Final pH	Free Chlorine mg per ℓ	Virus Destruction
Chang et al.	Part. purif. Theiler's virus in tap water	25-27	6.5-7.0	4.0-6.0	98.6% in 10 min
		25-27	6.5-7.0	4.0-6.0	99% in 5 min
Neefe et al.	Feces-borne Inf. Hepat. virus in dist. water	Room	6.7-6.8	3.25	30 min cont. time protected all of 12 volunteers
Lensen et al.	Purif. Polio II in dist. and lake water	19-25	7.4-7.9	1.0-1.5	10 min cont. time protected all of 164 inoc. mice
Clarke and Kabler	Purif. Coxsackie A2 in demand-free water	3-6	6.9-7.1	0.58-0.62	99.6% in 10 min
		3-6	6.8-7.1	1.9-2.2	99.6% in 4 min
		3-6	6.9-7.1	3.8-4.2	99.6% in 2½ min
		3-6	8.8-9.0	1.9-2.0	99.6% in 24 min
		3-6	8.8-9.0	3.7-4.3	99.6% in 9 min
		3-6	8.8-9.0	7.4-8.3	99.6% in 5 min
		27-29	6.9-7.1	0.16-0.18	99.6% in 4 min
		27-29	6.9-7.1	0.44-0.58	99.6% in 3 min
		27-29	8.8-9.0	0.10-0.18	99.6% in 10 min
		27-29	8.8-9.0	0.27-0.32	99.6% in 7 min
		27-29	8.8-9.0	0.92-1.0	99.6% in 3 min
Weidenkopf	Purif. Polio I (Mahoney) in demand-free water	0	6.0	0.39	99.6% in 3½ min
		0	6.0	0.80	99.6% in 1½ min
		0	7.0	0.23	99.6% in 8 min
		0	7.0	0.53	99.6% in 4½ min
		0	8.5	0.53	99.6% in 16 min
		0	8.5	1.95	99.6% in 7½ min
		0	8.5	5.00	99.6% in 3 min
Kelly and Sanderson	Purif. Polio I (Mahoney) in demand-free water	25-28	7.0	0.21-0.30	99.9% in 3 min
		25-28	9.0	0.21-0.30	99.9% in 8 min
	Purif. Polio III (Saukett) in demand-free water	25-28	7.0	0.11-0.20	99.9% in 2 min
		25-28	9.0	0.11-0.20	99.9% in 16 min

TABLE 78 (continued)

Investigator	Virus	Temp. °C	Final pH	Free Chlorine mg per ℓ	Virus Destruction
Kelly and Sanderson (cont'd)	Purif. Coxsackie B5 in demand-free water	25-28	7.0	0.21-0.30	99.9% in 1 min
		25-28	9.0	0.21-0.30	99.9% in 8 min
		1-5	7.0	0.21-0.30	99.9% in 16 min
		1-5	8.0	0.21-0.30	99.9% in 30 min
Clarke et al.	Purif. Adenovirus 3 in BOD demand-free water	25	8.8-9.0	0.20	99.8% in 40-50 sec
		25	6.9-7.1	0.20	99.8% in 8-16 sec
		4	8.8-9.0	0.20	99.8% in 80-100 sec
		4	6.9-7.1	0.20	99.8% in 8-10 sec

sufficiently large dose, a low residual chlorine concentration will remain in the water after treatment, providing continued disinfectant action. This prevents regrowth and protects against accidental contamination during distribution.

Despite chlorine's widespread use and some reports of effective virus kill, other reports are less optimistic about its performance against virus. Clarke et al. (129) and Sobsey (600) reported the isolation of viruses in chlorinated drinking water in Paris (1 pfu/300ℓ) and South Africa (1 pfu/10ℓ). In view of the fact that only one to two viruses of some types are sufficient to cause infection, anything less than 100 percent inactivation may be unacceptable. But with chlorine, even the absence of any "living" viruses still may not be acceptable. McClanahan (429) reported that chlorine removes the protein coat of a virus - thus rendering the virus technically nonviable - but may leave the infectious nucleic acid core intact. Consequently, a water supply free of any living viruses may still be infectious.

Ozone is equally as effective as chlorine against bacteria and viruses and has a much faster reaction rate. Once the proper "threshold" ozone dose is applied (usually less than 5 mg/ℓ), the bactericidal action is almost instantaneous. Tests have shown ozone to be between 600 and 3,000 times more rapid than chlorine in its destruction of bacteria (429). McClanahan (429) was unable to recover viable nucleic acids from ozonated water, suggesting that virus destruction was complete, as opposed to the action of chlorine. Venosa (660) reported that protozoal cysts resistant to chlorine were easily inactivated by ozone. Furthermore, the biocidal character of ozone is not affected by pH, as is the biocidal character of chlorine.

Ozone is not without problems; however, ozone leaves no residual. It has a fairly short half-life in water and rapidly loses all disinfectant ability. Experience in Europe has revealed few problems along these lines, but the added margin of safety with chlorine has worked against the adoption of ozone in the United States. The versatility of ozone and its relatively greater disinfecting ability has led to the suggestion that initial ozonation could be followed by low-level chlorination to provide a residual. However, little research has been conducted along these lines.

Murphy (469) and Vajdic (656) both reported that gamma radiation was as good a disinfectant as chlorine against bacteria and was somewhat better against the more chlorine-resistant biologicals. Ultraviolet is a proven bactericide, but research on other biocidal characteristics has been limited. Nevertheless, any radiation treatment suffers from operational difficulties, and, like ozone, provides no residuals.

The sanitation districts of Los Angeles County are in the final stages of an extensive study for EPA and the California State Water Resources Board. This study titled "The Pomona Virus Study," has evaluated virus removals by various combinations of tertiary treatment processes and followed by chlorination or ozonation. Their conclusions in part are that the majority of virus inactivation occurs during disinfection and the main function of the preceeding tertiary unit processes was that of removing substances (turbidity, organics, etc.) which interfere with efficient disinfection. In virus seeding experiments involving combined chlorine residuals of 5 mg/ℓ average seed virus removals of 4.7 to 5.1 logs were achieved. With 10 mg/ℓ of combined chlorine residuals, 5.2 logs of virus removal were achieved. In seeding experiments involving ozonation, average log virus removals ranged from 5.1 to 5.4 logs.

SECTION 10

ADVANCED WATER TREATMENT: ADSORPTION ONTO ACTIVATED CARBON AND OTHER MATERIALS

INTRODUCTION

Activated carbon adsorption (or simply carbon adsorption) is employed to remove color, odor, taste, and refractory organic compounds from water. Many water treatment plants presently pass their effluent through a carbon column or fine-grain carbon bed to polish the final product. Available data indicate that carbon adsorption is an effective method for removing synthetic and natural organic contaminants, particularly chlorinated hydrocarbons and organophosphorus pesticides, from water. Carbon adsorption may also be used to remove some metals. There is some adsorption of the free metal, but metal removal can be greatly enhanced by the addition of an organic chelating agent prior to passage through the carbon. The carbon will readily adsorb the chelating agent, thereby also removing the complexed metal.

The literature that has been reviewed on the effectiveness of the adsorption process in potable water treatment is summarized in Table 79. The major portion of the research to date has involved tertiary wastewater treatment application, although there has been substantial work also in the water treatment field as shown in Table 79. With the recent concern over residual organics in U.S. water supplies, one can anticipate increased activity in both pilot and full-scale activated carbon systems for treating water supplies.

In addition to activated carbon, synthetic polymeric adsorbents have been extensively tested and show promise for potable water treatment. They are not widely used in water treatment plants but have been tested in pilot-scale installations. Some tests have indicated higher removal efficiencies for synthetic adsorbents than for activated carbon for some contaminants. Inorganic adsorbents, such as clays and magnetite, are also capable of contaminant removal.

WATER QUALITY PARAMETERS

Activated carbon adsorption was used following chemical coagulation and rapid sand filtration at the much-publicized Windhoek, South Africa, water treatment plant. The influent

TABLE 79. LITERATURE REVIEWED PERTAINING TO ADSORPTION

Contaminant	Reference Number
Water Quality Parameters	
Ammonia	442, 610
BOD	515, 610
COD	515
Chlorides	442, 551, 589
Nitrates	442, 610
Nitrites	610
Oil and grease	316
Phosphates	515
Sulfates	610
Sulfides	378
Suspended solids	515
Taste and odors	70, 157, 276, 282, 316, 378, 442, 524
Turbidity	70, 276, 442
Elemental Contaminants	
Arsenic	467, 506
Barium	506
Cadmium	467
Chromium	506
Iron	442
Lead	467
Mercury	157, 309, 394, 467, 506, 640, 641
Selenium	506

TABLE 79 (continued)

Contaminant	Reference Number
Elemental Contaminants	
Silica	126
Biocidal Contaminants	
DDT	157, 192, 537, 546, 589
DDE	192
Aldrin	157, 192, 537
Dieldrin	157, 192, 316, 537, 546
Endrin	157, 192, 537, 546
Carbamates	193
Chlorinated hydrocarbons	157, 192, 316, 537, 551, 589
Organophos	157, 192, 467, 537
Herbicides	157, 467
Lindane	546
Other (general)	157, 262, 546, 570
Synthetic/Organic Contaminants	157, 262, 276, 284, 316, 352, 442, 455, 467, 506, 537, 589, 610, 632
Biological Contaminants	
Polio virus	467
Virus	467, 498

to this plant was treated wastewater which was subsequently mixed with surface water for direct reuse. Stander and Funke (610) reported on the effluent quality through the pilot plant. The concentration of ammonia nitrogen in the effluent was lowered from 0.3 to 0.1 mg/ℓ by passage through an activated carbon filter. ABS and BOD were also significantly reduced, 82 and 67 percent, respectively.

Table 80, from Medlar (442), summarizes water quality analysis data from several water treatment plants in New England employing granular activated carbon filters. Two of these plants (Amesbury and Scituate) use carbon for both filtration and adsorption, while the remaining four use carbon for adsorption only. At each plant, the carbon significantly reduced ammonia levels, as between 33 and 100 percent of the influent ammonia was removed.

Phillips and Shell (515) presented a study of the effectiveness of granular activated carbon and other general contaminants in removing BOD. The study was conducted at a pilot water plant treating wastewater effluent by chemical coagulation, filtration, and passage through 16-ft carbon columns. Data are presented in Table 81. BOD removal by the carbon columns averaged 33 percent, while COD was reduced 80 percent.

TABLE 81. ACTIVATED CARBON FILTRATION AT COLORADO SPRINGS PILOT PLANT (515)

	Influent	Effluent	Removal
	(mg/ℓ)		(%)
BOD_5	3	2	33
COD	41	8	80
SS	4	3	25
PO_4	2	1	50
ABS	0.9	0.03	97

Activated carbon is highly efficient for removing non-colloidal, soluble, aromatic-structured color sources. David Volkert and Associates (157) indicated that the carbon removal efficiency for color-producing substances is 100 percent of methylene-blue active substances. Table 80 shows that the color removal efficiency of carbon filters in six water treatment plants was nearly 100 percent (Medlar, 442). Activated carbon has also been used industrially for decolorizing organic dye waste effluents.

TABLE 80. SUMMARY OF WATER QUALITY ANALYSIS DATA FROM ACTIVATED GRANULAR CARBON (442)

		Amesbury Mass.	Newburyport Mass.	Scituate Mass.	Somerset Mass.	Manchester N.H.	Burlington Mass.
Color (APHA):	Raw	115	50	250	30	20	34
	Finished	0	0	5	2	0	0
Turbidity (N.T.U.):	Raw	14.5	2	4	1.8	1	1.0
	Finished	0.2	0	0.12	.13	0	0.2
Iron (mg/ℓ):	Raw	3.5	0.25	1.57	.35	0.4	0.51
	Unfinished	0.03	0.16	0.05	.05	trace	0.10
Odor (ton):	Raw	3	2	3	1	1	3
	Unfinished	0	0	0	0	0	0
Specific Conductance (μmhos/cm):	Raw	155	102	150	76	--	190
	Unfinished	230	280	268	107	--	235
Nitrate (mg/ℓ):	Raw	0.0	0.0	0.2	0.1	--	0.4
	Unfinished	0.0	0.5	0.2	0.2	--	0.4
Ammonia (mg/ℓ):	Raw	0.7	.02	0.22	.07	--	0.21
	Unfinished	0.0	.01	0.13	.02	--	0.14
Silica (mg/ℓ):	Raw	11.0	4.6	7.7	.7	--	
	Unfinished	9.6	5.4	8.3	.8	--	

Recently, the nonionic polymeric adsorbents, such as Amberlite XAD-7, have been gaining popularity for this purpose (589).

While Table 80 and the operation at Windhoek (610) show that both organic and ammonia nitrogen were reduced by carbon adsorption, they also show that oxides of nitrogen (nitrate and nitrite) were not reduced.

Oils derived from natural, domestic, or industrial sources may occasionally be found in wastewater effluents and in water supplies. Hyndshaw (316) reported that petroleum products are very quickly adsorbed from water by activated carbon. He stated that for emergencies, such as gasoline or oil spills, large quantities of powdered activated carbon will remove the hydrocarbons.

Reduction of phosphates as PO_4 from 2 mg/ℓ in the carbon filter influent to an effluent concentration of 1 mg/ℓ is shown in Table 81 (515). Sulfate as SO_4 was not reduced from its concentration of 220 mg/ℓ by carbon filtration at the Windhoek plant. However, sulfides in the form of hydrogen sulfide, and polysulfides that result from the reaction of chlorine with hydrogen sulfide were removed from well water, according to Lee (378). The removal was indirectly evidenced by the destruction of sulfide odor in the water supply and resulted from a catalytic reaction with the carbon rather than from adsorption.

Activated carbon is commonly used for removing tastes and odors from water. Undesirable odors in water are caused by the vapors from many chemicals, including halogens, sulfides, ammonia, turpentine, phenols and cresols, picrates, and various hydrocarbons and unsaturated organic compounds, some of which have not been identified. Tastes and odors are also caused by substances produced by living microorganisms or decaying organic matter. Some inorganic substances, such as metal ions in high concentrations (especially iron) also impart taste and odor to water. Removal of many tastes and odors with activated carbon approaches 100 percent. Successful taste and odor removal from a municipal water supply by carbon bed filters was demonstrated in Buckingham, England (Hager, 276). The water was taken from a river that runs through farmland and had an earthy taste that resisted chlorination; clarification and sand filtration; and experimentation with ozone, permanganate, and chlorine. At Goleta, California, taste and odor in the water supply were also eliminated by passage through granular activated carbon beds (276). Popalisky and Pogge (524) reported that powdered activated carbon was used at a water plant treating Missouri River water to eliminate taste and odor caused by microorganic compounds. The compounds and their concentrations were not identified.

Hansen (282) reported that installation of granular carbon filters at Mount Clemens, Michigan, completely removed the septic and phenol tastes and odors that were present in the Clinton River water supply during runoff periods. Granite City, Illinois, reported a similar experience (70). The dosage of activated carbon required to remove taste and odor is influenced by chlorine application. Generally, carbon should be applied before chlorination (316).

Many pesticides and herbicides produce tastes and odors when present only in very small concentrations in water. Hyndshaw (316) used the beta isomer of benzene hexachloride to illustrate the influence of a small concentration of a biocide on the taste and odor quality of water. This substance, when present in amounts as little as 17 ppb in odor-free water, gives a threshold odor number (TON) of 8. (The threshold number denotes the number of volumes of odor-free water required to dilute the odor concentration to a point where the odor is just barely perceptible.) Other organic pesticides and their concentrations and subsequent effects upon taste and odor are shown in Table 82 (316).

TABLE 82. ODOR IMPARTED TO ODOR-FREE WATER BY PESTICIDES AND HERBICIDES (316)

Substance	Concentration ppm	Odor TON
Toxaphene	0.84	6
2,4 D (isoctyl)	2.0	17
2,2 D	92.5	4
D-D	0.0235	17
Rothane	50.0	Nil
Chlordane	0.07	140
BHC	0.0175	8

A simple study employing the threshold odor test gave evidence of the reduction of these compounds by activated carbon. The amount of activated carbon required to reduce the odor of each compound to a palatable level is presented in Table 83 (316).

TABLE 83. ACTIVATED CARBON REQUIRED TO REDUCE ODORS CAUSED BY PESTICIDES AND HERBICIDES TO PALATABLE LEVELS (316)

Substance	Concentration of substance ppm	Odor TON	Carbon Dosage ppm	Odor after Carbon TON
Parathion	10	50	20	4
37 gamma BHC	25	70	15	1.4
Malathion	2	50	20	4
2,4 D	6	50	40	3
DDT	5	70	4	3

Significant reduction in turbidity and suspended solids concentration is also effected with activated carbon. The data in Table 80 show that turbidity is reduced both at plants where granular activated carbon is used for filtration and adsorption and where it is used for adsorption only. Passage of water through 16-ft carbon columns effected 25 percent removal of suspended solids at Colorado Springs (Table 81) (515).

Turbidity removal is essentially complete at Nitro, West Virginia, where activated carbon has replaced sand in the filters (276). Final effluent JTU's are typically less than 0.1. At Granite City, Illinois, Blanck and Sulick (70) report that suspended solids removal by carbon filtration exceeds that achieved with sand filters.

ELEMENTAL CONTAMINANTS

Although little data from municipal water purification applications are available, it appears that activated carbon can provide some removal of heavy metals. Direct adsorption provides some removal, but efficiencies can be increased to nearly 100 percent by adding an organic chelating agent (537). The carbon removes the complex by adsorbing the organic agent, removing the metal along with it.

Patterson (506) cited evidence that filtration of water containing 0.2 mg/ℓ arsenic through a charcoal bed yielded an effluent containing 0.06 mg/ℓ of arsenic, or 70 percent removal. He cited another report of 40 percent reduction of arsenite by activated carbon from an initial concentration of 0.5 mg/ℓ to a concentration of 0.3 mg/ℓ. Morton and Sawyer (467) tested heavy metal adsorption during filtration through a granular bed of attapulgite clay. Data are presented in Table 84 that show the amounts of metals remaining in the effluent after filtration of various quantities of water at two different rates. The initial solutions contained 1 mg/ℓ of each metal. Arsenic was reduced from 0.97 and 0.98 mg/ℓ to 0.02 and 0.03 mg/ℓ upon filtration of 2.5 volumes of water per volume of bed.

TABLE 84. REMOVAL OF HEAVY METALS BY PERCOLATION WITH GRANULAR LOW VOLATILE MATTER ATTAPULGITE CLAY (467)

Ratio of Volume Percolate to Volume Bed	Recoveries in Effluent			
	As	Cd	Pb (ppm)	Hg
			Slow Rate*	
Initial Concentration	0.97	1.00	1.06	1.11
2.5	0.02	0.01	0.01	0.004
5.0	0.12	0.01	0.01	0.016
10.8	0.56	0.01	0.01	0.116
			Fast Rate†	
Initial Concentration	0.98	1.00	1.06	1.11
2.5	0.03	0.01	0.01	0.010
5.0	0.16	0.01	0.01	0.072
12.6	0.68	0.01	0.01	0.152

* 960 gal/ton clay/hour.
† 2,880 gal/ton clay/hour.

In a study cited by Patterson (506), carbon adsorption did not improve barium removal efficiency over that achieved with chemical coagulation and clarification. Patterson notes that others have also found poor removal efficiency for barium.

Cadmium removal data are also presented in Table 84 (467). Removal of 99 percent was achieved and was unaffected by the changes in flow volume and rate.

Some success has been reported from pilot plant work on chromate removal by activated carbon. Patterson (506) reported on a study of metal removal from municipal wastewater plant effluent, in which initial hexavalent chromium levels of 0.09 to 0.19 mg/ℓ were reduced to 0.04 mg/ℓ or less. The average effluent concentration reported was 0.017 mg/ℓ.

Initial hexavalent chromium levels of 5 mg/ℓ were reduced to 0.09 mg/ℓ following carbon adsorption. Patterson noted that it appears that activated carbon may not be equally effective at higher chromate levels.

Table 80 reports removal of iron at six water treatment plants by granular activated carbon filtration. Removal efficiencies ranged from 99 percent to 36 percent.

Lead removal is shown in Table 84 for adsorption onto attapulgite clay. Over 99 percent removal was achieved at all flow volumes and rates tested.

Mercury adsorption has been studied extensively. In Table 84 , it is shown that adsorption onto attapulgite clay of over 99 percent was achieved after filtration of 2.5 volumes of water per volume of bed. Removals decreased to 86 percent removal as 12.6 volumes per volume of bed were filtered. Logsdon and Symons (394) conducted jar tests using powdered activated carbon. Increasing carbon dosages increased removal of both inorganic and organic (methyl) mercury. Organic mercury removal was more efficient than inorganic mercury removal for a given dosage of powdered activated carbon at effluent concentrations less than 2 ppb. Residual concentrations of 0.8 ppb of inorganic and 0.2 ppb of methyl mercury were achieved. Further tests were performed in which powdered carbon was added just before alum coagulation to improve mercury removal. Removal by alum alone was about 40 percent, whereas removal with 65 mg/ℓ of activated carbon plus 30 mg/ℓ of alum was in excess of 70 percent with an initial mercury concentration of 9.3 ppb.

The capacity of granular activated carbon for removing mercury from water was also evaluated by pumping mercury solutions through columns (394). Average influent concentrations ranged from 20 to 29 ppb. Mercury removals declined as the number of bed volumes (based on gross void space) of water treated increased. Columns with 3.5-min contact times removed 80 percent of the inorganic mercury for up to 15,000 bed volumes of water and 80 percent of the methyl mercury for up to 25,000 bed volumes treated. Evaluation of carbon column performance at 80 percent removal indicated that as contact time in the column increased, the amount of mercury adsorbed by the carbon increased. Also, granular activated carbon adsorbed more methyl mercury than it did inorganic mercury per gram of adsorbent. This behavior was expected, since activated carbon has a high capacity to adsorb organics.

Thiem (640) also conducted jar tests on mercury adsorption using powdered activated carbon. Solutions containing 10 ppb mercury were brought into equilibrium with various carbon dosages. Less than 30 percent of the mercury was adsorbed at a carbon dosage of 10 mg/ℓ at pH 7. When 100 mg/ℓ of carbon was applied to the test solution, removal approached 80 percent. Removal decreased with increasing pH, with best removal occurring at pH 7. The addition of chelating agents such as EDTA or tannic acid prior to contact with the carbon increased adsorption. Concentrations of as little as 0.02 mg/ℓ of EDTA or 1 mg/ℓ of tannic acid increased removals from 10 to 30 percent, depending upon the carbon dosage that was applied and the pH of the system.

Patterson (506) summarized the data on activated carbon removal of mercury, saying that the highest percentage removals (80 to 95 percent) are achieved with more concentrated mercury solutions. However, lowest effluent mercury results from treatment of less concentrated waters, although the relative efficiency is less. Thus carbon treatment of initial mercury below 1 ppb yields an efficiency of removal of less than 70 percent, but effluent mercury is below 0.25 ppb. Carbon treatment of initial mercury concentrations of 5 to 10 ppb yields about 80 percent removal and effluent levels of below 2 ppb. Carbon treatment of initial mercury concentrations between 10 and 100 ppb yields 90 percent or greater efficiency.

Poor selenium removal from well water by activated carbon has been reported in one case cited by Patterson (506). Removal was less than 4 percent. Poor or no reduction of silica has also been shown by the data from six operating water purification plants employing carbon adsorption and filtration (442).

BIOCIDAL CONTAMINANTS

As with other synthetic-organic compounds, some of the organic pesticides and herbicides that are resistant to removal by conventional treatment techniques are effectively removed by adsorption. David Volkert and Associates (157) cited evidence that over 99 percent of the following chlorinated hydrocarbons can be adsorbed by activated carbon:

DDT
Aldrin
Dieldrin
Endrin
Chlordane
Heptaclor epoxide
Lindane
Methoxychlor
Toxaphene

Laboratory studies cited by Stone (537) have shown that the following reductions in chlorinated hydrocarbon concentrations can be achieved by contacting with appropriate doses of activated carbon (Table 85):

TABLE 85. ACTIVATED CARBON REMOVALS OF CHLORINATED HYDROCARBONS ACHIEVED IN LABORATORY EXPERIMENTS (537)

Substance	Initial Concentration	Treated Concentration	Percent Reduction
Chlorinated hydrocarbons	6.3 ppb	0.04-0.11 ppb	98-99
DDT	5 ppm	-	90
DDT	5 ppm	-	76
Aldrin	6.6 ppb	-	90
Aldrin	5 ppm	-	85
Aldrin	-	-	99
Dieldrin	-	-	99
Dieldrin	0.5-10 ppb	0.25 ppb	50-97
Dieldrin	4.4 ppb	-	99
Endrin	5 ppm	-	86
Endrin	0.5-10 ppb	0.25 ppb	50-97
Chlordane	50 ppm	-	99
Lindane	10 ppb	1 ppb	90
Lindane	25 ppb	-	90
Lindane	1 ppb	0.05 ppb	95

Activated carbon removals of several pesticides are well illustrated by results of laboratory studies cited by the California State Water Resources Control Board (615) shown in Table 86.

TABLE 86. REMOVAL OF SPECIFIC TOXIC MATERIALS BY CARBON ADSORPTION (615)

Carbon	Residual (ppb)						
Dosage (mg/ℓ)	Aldrin	Endrin	Dieldrin	DDT	DDD	DDE	Toxaphene
Control	48	62	19	41	56	38	155
1.0	--	--	--	41	--	34	147
2.0	26	15	6.3	--	6.9	--	80
2.5	--	--	--	21	--	29	--
5.0	15	3.4	2.4	3.7	3.7	12	31
10.0	12	1.5	1.1	--	2.2	--	2.7
12.5	--	--	--	<1	--	3.3	--
25.0	6.3	0.56	--	--	0.45	1.1	--
50.0	4.4	0.22	--	--	0.35	0.9	--

Adsorption of the carbamate pesticides Sevin and Baygon on granular activated carbon was investigated by El-Dib et al. (193). Passage of a 5 mg/ℓ solution of Baygon through carbon columns effected complete removal for up to 273 bed volumes of water when a contact time of 3.76 min was allowed. In the case of Sevin, 1,800 bed volumes were passed with complete removal under the same conditions. Rapid breakthrough of the pesticides into the effluent occurred, however, as the contact times were decreased to 1 or 0.5 min.

According to reference 570, the available data on organophosphorus pesticide removal indicates that the efficiency of activated carbon ranges from 50 to over 99 percent. Stone and Company (537) cited laboratory-scale tests showing that Parathion was reduced from 10 to 2.5 mg/ℓ using 20 mg/ℓ powdered carbon; and from 11.4 to 0.05 mg/ℓ using dual granulated carbon filters. Malathion was reduced in laboratory-scale tests from 2 to 0.25 mg/ℓ with 10 mg/ℓ powdered carbon. Some data on removal efficiencies of the organic herbicides were also cited. They report that over 99 percent removal of 2,4-5-T and 2,4-D is possible.

Robeck et al. (546) surveyed the effectiveness of various water treatment processes in pesticide removal. Table 87 summarizes their results using carbon in both a slurry form and in beds.

TABLE 87. SUMMARY OF CUMULATIVE PESTICIDE REMOVAL AT 10-ppb LOAD (546)

Process	Pesticide Removed - Percent					
	DDT	Lindane	Parathion	Dieldrin	2,4,5-T Ester	Endrin
Carbon:						
Slurry						
5 ppm		30	> 99	75	80	84
10 ppm		55	> 99	85	90	90
20 ppm		80	> 99	92	95	94
Bed 0.5 gpm/cu ft	> 99	> 99	> 99	> 99	> 99	> 99

In addition to activated carbon, other adsorbents such as clays and synthetic polymeric adsorbents are capable of removing biocidal contaminants. Morton and Sawyer (467) studied the adsorption of diazinon, an organophosphorus pesticide, onto attapulgite clay. Coarse-ground, high-volatile matter attapulgite was stirred with contaminated water in laboratory experiments. At least 50 percent of the diazinon in a 0.1 mg/ℓ solution was removed by the clay in a 10 percent clay suspension. An investigation of the use of the synthetic polymer Amberlite XAD-4 for the removal of various pesticides was cited by Stone and Company (537). The pesticides examined included lindane, B-BHC, aldrin, and dieldrin in tap water at initial concentrations of 1 ppb each. Consistent removals of over 60 percent were reported.

Schwartz (570) also studied the adsorption of selected pesticides on activated carbon and mineral surfaces. He found that the clay minerals ilite, kaolinite, and montmorillonite suspended in dilute pesticide solutions adsorbed very little 2,4-D or isopropyl N-(3-chlorophenyl) carbamate (CIPC). Adsorption of CIPC from aqueous solution with powdered activated carbon, however, was extensive (>90 percent).

SYNTHETIC/ORGANIC CONTAMINANTS

Adsorption is commonly cited as a presently available technology for removing particulate, colloidal, and soluble organic contaminants from water. Many of the organics present in water supplies - particularly the soluble and colloidal organics - are of a refractory nature, i.e., they resist removal by conventional methods. A number of these are potentially toxic or carcinogenic and, as a result, their detection, identification, and treatment in water is receiving increasing attention. These substances, even in small amounts, contribute to taste and odor conditions and may pose a chronic health hazard. As has been discussed, activated carbon is widely applied for taste and odor removal; however, its effectiveness for removing residual organics has just come under study in recent years. The delay has been caused in part by the lack of standard procedures for identifying and classifying the vast assortment of organics that occurs in trace quantities in water (262,455). The delay is also due to the search for a gross organic parameter that can be used as a measure of organics. The more common parameters used include carbon chloroform extract (CCE), liquid extraction, paper and gas chromatography, fluorescent spectroscopy, and radiation.

Traditionally, carbon life expectancy has been based on the capability of the carbon to absorb tastes and odors. But research has shown that the life expectancy of carbon to reduce carbon chloroform extract or organic compounds is somewhat less than that to remove tastes and odors (442). Medlar (442) suggested that monitoring carbon chloroform extract concentration in carbon filtered water would provide a conservative estimate of filter performance, but noted that the CCE test may not encompass all the compounds that should be considered.

Carbon chloroform extract indicates the presence of stable organic compounds in water. The extract has an operational definition and is a mixture of organic compounds that can be adsorbed onto activated carbon and then desorbed with organic solvents under specific controls. Examples of substances measured with this method include substituted benzene compounds, kerosene, polycyclic hydrocarbons, phenylether, and insecticides. The efficiency of activated carbon in reducing CCE depends upon several factors including water temperature, initial amount of contaminant, and the molecular weight of the contaminant (157). Percentage removals must therefore be determined by laboratory testing. Removals ranging from 50 to 99 percent were reported by David Volkert and Associates (157).

The adsorption of polycyclic (polynuclear) aromatic hydrocarbons (PAH) from water by activated carbon was discussed by Harrison et al. (284). These compounds are potential carcinogens under certain conditions. Carbon adsorption has been shown to give 99 percent removal of PAH from water filtered by prior seepage through river bank soil.

Bis-ethers are synthetic organic compounds that may occur in water associated with industrial discharges. Stone and Company (537) cited laboratory tests of activated carbon treatment in which isopropyl ether concentrations were reduced from 1,023 to 20 mg/ℓ, butyl ether concentrations from 197 mg/ℓ to nil, and dichloroisopropyl ether concentrations from 1,008 mg/ℓ to nil.

The treatment of dilute phenolic industrial wastewater was reviewed by Patterson (506). Adsorption onto activated carbon has been employed to remove over 99 percent of the phenol present in process waters with initial concentrations ranging from 5,325 mg/ℓ to 0.12 mg/ℓ. Final phenol concentrations ranged from 0.25 mg/ℓ for treatment of a concentrated solution to 0.001 mg/ℓ for treatment of weaker solutions.

Foaming agents such as linear alkyl benzene sulfonate in concentrations up to 5 mg/ℓ can be removed by activated carbon with 90 to 100 percent efficiency according to evidence cited by David Volkert and Associates (157). Table 81 (Phillips and Shell, 515), which presents data on activated carbon filtration at the Colorado Springs pilot plant, shows 97 percent removal of alkyl benzene sulfonate (ABS). Stander and Funke (610) reported reduction of ABS from 4 to 0.7 mg/ℓ at the Windhoek pilot plant. Organic acids are also reported to have been reduced from 1 to 0.4 mg/ℓ.

Morton and Sawyer (467) studied the adsorption of two organic compounds - diethylstilbestrol (DES), which is a hormone and aflatoxin, which is a natural toxin produced by fungi - onto attapulgite clay. Attapulgite is a magnesium aluminum silicate clay that exhibits a high degree of adsorption for low-weight organic molecules. Coarse-ground, high-volatile-matter attapulgite was contacted with contaminated water in laboratory experiments. DES at a concentration of 5 ppb was decreased 68 and 76 percent by contacting with 1.1 and 10 percent (by weight) clay suspensions, while in a 50 ppb solution the removals were 68 and 89 percent, respectively. More than 98 percent of the aflatoxin at concentrations of 0.5 ppb and 5.0 ppb was removed by both 1.1 and 10 percent clay suspensions. The results of column percolation studies through granular low-volatile-matter clay are presented in Table 88.

TABLE 88. REMOVAL OF ORGANICS BY PERCOLATION WITH GRANULAR, LVM ATTAPULGITE

Ratio of percolate volume to bed volume	Recoveries in Effluent	
	Slow rate*	Fast rate†
Initial Concentration	52 ppb	48 ppb
2.5	ND‡	ND
5.0	ND	1
10.8	ND	--
12.6	--	2
Initial Concentration	17 ppb	17 ppb
2.5	ND	ND
5.0	ND	ND
10.8	ND	--
12.6	--	ND

* 960 gal/ton clay/hour.
† 2,880 gal/ton clay/hour.
‡ ND = not detectable.

Stone and Company (537) cited data concerning the treatment of polychlorinated biphenyls (PCB's) by adsorption onto clay minerals and Amberlite polymeric adsorbents. In laboratory tests, Amberlite XAD-4 removed up to 76 percent of the PCB present in solutions. Several clay minerals demonstrated PCB removal capability in laboratory tests: illite - 60 percent, montmorillonite - 40 percent, and kaolinite - 40 percent. Kinoshita and Sunata (352) evaluated the adsorption of PCB onto powdered activated carbon in a jar test and found that the initial concentration of 100 ppb PCB was reduced to 10 ppb in the product water.

The Amberlite adsorbents represent a new technology for adsorbing organic molecules from water. They are used specifically for adsorbing aromatic and aliphatic compounds. According to Simpson (589), small molecules such as phenol are effectively adsorbed by Amberlite XAD-4, while for a larger molecule such as an alkylbenzene sulfonate, Amberlite XAD-2 has a much higher adsorptive capacity. Phenol is a model aromatic, low-molecular weight compound that is considered to be highly objectionable, as are some of the chlorinated phenol products. Using the XAD-4 adsorbent, up to 40 bed volumes of a 500 ppm water solution of phenol were treated with less than 10 percent leakage at a flow rate of 0.5 gal/ft^3/min. At a higher flow rate of 2.0 gal/ft^3/min, 20 bed volumes were treated with less than 10 percent leakage. It was further found that the adsorptive capacity was higher for chlorinated phenols than for simple phenol. Simpson (589) cited a laboratory study in which the removal efficiency of Amberlite XAD-2 for a list of organics at flow rates of 1.25 gpm/cu ft were determined. The results are presented in Table 89. Nonionic compounds were removed with 100 percent efficiency while ionized compounds were less effectively removed.

In the nationwide study of water supplies and water treatment facilities by Symons et al. (632), it was concluded that both powdered and granular activated carbon treatment significantly reduced the trace concentrations of total trihalomethane in the product water.

TABLE 89. ADSORPTION OF ORGANIC COMPOUNDS ONTO AMBERLITE XAD-2 POLYMERIC ADSORBENT (589)

	Influent	Effluent	Retention Efficiency, %
1. Aliphatics			
a) alcohol: n hexanol	200 ppm	30 ppm	85
b) ester: ethyl butyrate	100	0	100
c) ketone: methylisobutylketone	100	0	100
2. Aromatics			
Benzene	100	0	100
Benzene sulfonic acid	3.0	2.1	31
p-toluene sulfonic acid	9.0	6.9	23
Benzoic acid	1.0	0.8	23
Benzoic acid (pH 3.2)	1.0	0	100
Phenol	0.4	0.22	45
Phenol (Amberlite XAD-7)	0.4	0.06	86
O-Cresol	0.3	0	100
2, 4-dimethyl phenol	0.4	0	100
p-nitrophenol	0.2	0	100
2-methylphenol	0.3	0	100

TABLE 89. (continued)

2. Aromatics (continued)	Influent	Effluent	Retention Efficiency, %
4, 6-dinitro-2-aminophenol	0.4	0.22	43
Phenylenediamine	0.9	0.02	98
Aniline (Amberlite XAD-7)	4.0	0	100
Naphthalene	0.05	0	100
2-hydroxy-3 naphthoic acid	0.6	0.37	39

BIOLOGICAL CONTAMINANTS

The limited efficiency of activated carbon in removing viruses from wastewater was discussed in the advanced wastewater treatment section of this report. Results show that activated carbon is inefficient in removing viruses from drinking water. Oza and Chaudhuri (498) suggested that the inefficiency may be due to the exclusion of viruses from the micropores of activated carbon because of their size. Coal adsorption of a bacterial virus was investigated, and results indicated that coal may be a more effective adsorbent than activated carbon. Morton and Sawyer (467) demonstrated that attapulgite clay also has the capacity to adsorb polio virus from water. Aqueous virus-clay suspensions were shaken for 1 min and then filtered. High-volatile-matter clay completely removed virus infectivity from a 20 percent clay suspension with an initial virus concentration of 16 million infectious particles per mℓ. However, reducing the contact time from 30 to 5 min or the clay concentration from 20 to 5 percent resulted in incomplete removal.

ADVANCED WATER TREATMENT: ION EXCHANGE

Ion exchange has its greatest current application in industrial and small-scale potable water supply operations. The most common use of ion exchange is for removal of hardness (calcium and magnesium cations) from municipal, industrial, household, and laboratory water supplies. It is particularly suited for desalting brackish water, pretreating water that must be almost completely demineralized for industrial use, and removing metals from industrial metal plating rinse wastewaters.

No one ion exchange resin is capable of removing all ionic contaminants. Various resins, depending upon their chemical nature, show preferential selectivity for specific ions. Table 90 presents data on ion selectivity for various types of exchange resins (506).

TABLE 90. ION EXCHANGE RESINS SELECTIVITY (506)

Resin	Resin Selectivity*
Strong-acid cation (Sulfonic)	Li^+, H^+, Na^+, NH_4^+, K^+, Rb^+, Cs^+, Mg^{+2}, Zn^{+2}, Cu^{+2}, Ca^{+2}, Pb^{+2}
Weak-acid cation (Carboxylic)	Na^+, K^+, Mg^{+2}, Ca^{+2}, Cu^{+2}, H^+
Strong-base anion (Type I)	F^-, OH^-, $H_2PO_4^-$, HCO_3^-, Cl^-, NO_2^-, HSO_3^-, CN^-, Br^-, NO_3^-, HSO_4^-, I^-
Strong-base anion (Type II)	F^-, $H_2PO_4^-$, OH^-, HCO_3^-, Cl^-, NO_2^-, HSO_3^-, CN^-, Br^-, NO_3^-, HSO_4^-, I^-
Weak-base anion	F^-, Cl^-, Br^-, I^-, PO_4^{-3}, NO_3^-, CrO_4^{-2}, SO_4^{-2}, OH^-

*Increasing selectivity left to right.

The more strongly a resin adsorbs a particular ion, the more complete the ion removal. However, a high affinity between a particular resin and a specific ion also means greater difficulty in regenerating the resin; that is, it is more difficult to release the adsorbed ions to make the resin reuseable. As a result, regeneration of an effective resin is seldom carried to completion, and the operational capacity of an ion exchange resin may be reduced to 50 to 60 percent of theoretical capacity.

Ion exchange processes are very sensitive to both clogging and fouling. An ion exchange resin bed is a good filter; therefore, suspended solids in water will clog the bed. Fouling results when the resin adsorbs materials which, because of their adsorption or absorption into the resin pores, cannot be removed in the regeneration step. Fouling often results from the irreversible sorption of high molecular weight organic acids.

The nonselective nature of most exchange resins is a drawback when attempting to remove a low-level contaminant from water. The simultaneous removal of other nontarget ions rapidly increases the cost of using the process. More selective resins are currently becoming available (Koerts, 359).

In the past, application of ion exchange processes was confined to the removal of ionic contaminants. Recently, however, some ion exchangers have been developed that can remove nonionic species, e.g., Al_2O_3, Fe_2O_3, H_2SiO_3, etc. Resinous adsorbents are also available that are particularly suited for removing organic compounds - including biocidal and synthetic/organic contaminants - from water. While often used in conjunction with true ion exchange processes, the mechanism of organic removal is actually adsorption; therefore, the resinous or polymeric adsorbents (as they are called) are discussed in the carbon adsorption section of this report. The literature utilized during this review on the removal of various contaminants by ion exchange methods is indicated in Table 91.

WATER QUALITY PARAMETERS

Reduction of total dissolved solids may be an important application of the ion exchange process when reclaimed wastewater for potable reuse is the objective of treatment. High total dissolved solids concentrations, from 500 to over 1,000 mg/ℓ, are found in wastewaters. Approximately 300 mg/ℓ of total dissolved solids is generally considered the increment added during one cycle of domestic use of a water supply. Ion exchange treatment of water and wastewater for removal of dissolved solids is most successful after prior treatment using both conventional and advanced techniques has taken place. This prevents clogging and fouling of the exchange resins.

Ion exchange is technically capable of producing a water with only 0.055 micromhos (μmhos) of specific conductance. One micromho will normally indicate a dissolved solids concentration of 0.5 to 0.6 mg/ℓ. However, water of such purity is rarely

TABLE 91. LITERATURE REVIEWED PERTAINING TO ION EXCHANGE

Contaminant	Reference Number
Water Quality Parameters	
Ammonia	101, 505, 506, 524
Chlorides	157, 505, 506, 524
Color	157
Cyanides	157, 506
Fluorides	157, 506
Hardness	505
Nitrates	63, 90, 101, 119, 157, 267, 297
Phosphates	63, 119
Sodium	119, 157, 505
Sulfates	119, 157, 505
Total dissolved solids	63, 119, 505, 506
Elemental Contaminants	
Arsenic	99, 506, 576
Barium	506
Boron	506
Cadmium	506
Chromium	506
Copper	359, 506
Iron	12, 506
Manganese	12, 506
Mercury	359, 506
Nickel	506

TABLE 91 (continued)

Contaminant	Reference Number
Selenium	506
Zinc	359, 506

required, and the costs to achieve such purity would be prohibitive. Complete demineralization of municipal water supplies is unwarranted and may even have adverse consequences, as the body requires certain trace concentrations of many minerals.

The EPA and the LA County Sanitation Districts jointly funded an advanced wastewater treatment facility in Pomona, California. The results of the ongoing tests have been comprehensively reported by Parkhurst (505) and Chen (119). The ion exchange system, which was proven to be of economic and technical feasibility, effectively reduced the total dissolved solids by 90 percent. The treatment involved passage first through a primary cation column and an anion column, then through a secondary cation column and a secondary anion column. A series of 80 complete operating cycles including exchange, backwashing, and regeneration was completed at a 2.5-gpm flow rate. The total dissolved solids concentrations of the feed and product waters averaged 610 mg/ℓ and 72 mg/ℓ, indicating an 89 percent average reduction. A complete listing of the various constituents at successive stages of the demineralization sequence is presented in Table 92 (505).

TABLE 92. AVERAGE WATER QUALITY CHARACTERISTICS OF THE ION EXCHANGE PILOT PLANT UNDER TYPICAL OPERATING CONDITIONS (505)*

	Carbon Column Effluent (feed)	Primary Cation Effluent	Primary Anion Effluent	Secondary Cation Effluent	Secondary Anion Effluent (product)
Calcium	53†	2.0	1.7	1.1	0.60
Magnesium	17	0.59	0.56	0.38	0.00
Sodium	126	61	59	16	15
Potassium	14	7.3	7.1	1.9	1.9
Ammonia, as N	20	9.6	9.2	4.0	3.8
Sulfate	72	72	3.6	3.6	1.3
Nitrate, as N	2.9	2.8	1.6	1.5	0.35
Chloride	135	132	84	83	14
Orthophosphate, as PO_4	27	27	15	14	0.25
Total alkalinity, as $CaCO_3$			51		39
pH	7.4	2.7	5.7	2.8	5.8
Conductivity	10	9.7	6.8	5.5	3.7
Silica, as SiO_2	23				23

*Data taken from May 1968 through December 1968.

†All constituents in mg/ℓ except: (1) pH and (2) conductivity (μmhos/cm).

All of the major anions and cations were removed to a considerable degree except silica. Silica removal can be accomplished, however, with highly basic anion resins (Betz, 63). Calcium and magnesium, the cations responsible for water hardness, are almost completely removed in the primary cation exchanger; but only about half of the sodium, potassium, and ammonium ions are removed in this column. The secondary cation exchanger, however, efficiently removed the majority of the remaining ions. Similarly, the primary anion exchanger removed most of the sulfate ions, while the nitrate, chloride, and orthophosphate ions were partially removed by both the primary and secondary stages.

David Volkert and Associates (157) cited evidence that the sodium and fluoride concentrations of water can be reduced by 95 percent using ion exchange. It reported that chloride, sulfate, and nitrate can be reduced by as much as 95 percent, depending on the degree to which the exchange resin can be regenerated.

The application of ion exchange to nitrate reduction of reclaimed wastewaters may be particularly important, as these waters frequently may contain nitrate concentrations in excess of the 10 mg/ℓ limit set by the U.S. Environmental Protection Agency as an interim primary drinking water standard. The use of ion exchange resins for the removal of ammonium and nitrate ions has been discussed in the adsorption section of this report. Few specific exchange resins are available for removal of the nitrate ion from wastewater; ammonium ion removal with certain specific zeolite resins can be quite effective. In wastewater treatment, significant nitrogen removal may be effected by removing ammonia with ion exchange; in water purification treatment, nitrate is often the most significant form of nitrogen. Strong nonspecific anion exchange resins have been applied to the removal of nitrates from drinking water; but because the resins are nonspecific, competition from sulfides, chlorides, silicates, and phosphates limits the target removal of nitrates.

This problem of competing ions may be significant when considering potable reuse, since reclaimed wastewaters that are high in nitrates are also likely to be high in these anions. The caking and clogging problems caused by iron, turbidity, and colloidal matter are also significant. In other words, ion exchange technology for nitrate removal is still developing. A successful application of the available technology has taken place at Long Island, New York (267,297). A few years ago, Long Island communities began experiencing rising nitrate levels in their drinking water supply source. Drinking water was obtained from groundwater wells that had been contaminated with nitrates from septic tanks and cesspools. An ion exchange process originally developed to demineralize industrial process water was adapted in a prototype ion exchange plant; it reduced the nitrate content of several thousand gallons of the Long Island water from 22 ppm to 0.5 ppm nitrate. A plant was then built that successfully provided ion exchange treatment for the Long Island well water under the constant flow and pressure requirements of a municipal water system. However, the well water was of relatively low total dissolved solids, and it is uncertain how this process would perform on waters of poorer quality.

A water quality parameter that is of interest in potable water treatment is color. According to David Volkert and Associates (157), ion exchange resins have been developed that will completely remove color-causing organic dye wastes, humates, and lignates.

ELEMENTAL CONTAMINANTS

Ion exchange techniques have been applied to the treatment of industrial process water containing trace metals for several years. Application of these techniques to drinking waters containing trace concentrations of these metals is analogous. In a review of the literature, David Volkert and Associates (157) found that several trace elements can be removed from water to a level of 95 percent. Table 93 lists these elements and gives the maximum concentrations that can be reduced to 1974 Standards and Guidelines in a single pass through an ion exchange process (157). Water containing concentrations of a particular contaminant higher than the maximum listed in the table can be either passed through a series of ion exchange columns with different types of resins, or pretreated by a method such as lime coagulation to precipitate the major amount of the metal present.

Calmon (99) noted that anion exchange treatment can be used to remove residual arsenic after lime coagulation is used to precipitate the major amount present. Both weak and strong-base ion exchange resins appear effective in removing arsenate and arsenite from drinking water (Patterson, 506). Calmon (99), treating an arsenate water containing 68 mg/ℓ arsenic at pH 6.95 with a weak-base anion exchange resin (Ionac A-260), reported 82 to 100 percent removal. Medium and strong-base resins (Ionac A-300, A-540, and A-550) were less effective.

Again using the weak-base anion exchange resin (Ionac A-260), Shen (576) treated water containing 106 mg/ℓ of arsenic. Only 20.7 percent of the arsenic was removed. When well waters with naturally high arsenic levels were treated, essentially 100 percent removal was achieved. The disparity of the results was not explained.

Bone char and activated alumina readily remove arsenic via an ion exchange mechanism. Arsenic sorption on bone char results in an irreversible change in the chemical structure of

TABLE 93. REMOVAL OF TRACE ELEMENTAL CONTAMINANTS FROM WATER BY ION EXCHANGE (157)

Contaminant	% Removal	Maximum* Concentration
Arsenic	95	2.0 mg/ℓ
Barium	95	20.0
Cadmium	95	0.2
Chromium	95	1.0 mg/ℓ
Copper	95	20.0
Cyanides	95	4.0
Lead	95	0.1
Iron	95	6.0
Manganese	95	0.1
Mercury	95	0.04
Selenium	95	0.2
Silver	95	0.1
Zinc	95	100

*Maximum concentration in raw water, which can be reduced to 1974 Standards and Guidelines in a single pass through process. If raw water concentrations are higher, then combination or duplication of processes or other processes must be considered.

the char. Consequently, exhausted bone char must be discarded; it cannot be regenerated. Activated alumina is regenerable.

Effective removal of barium by ion exchange has been reported. Patterson (506) cited a 98.5 percent reduction of barium from 11.7 to 0.17 mg/ℓ in a full-scale ion exchange groundwater softening plant in which a general nonspecific water softening resin was used.

According to Patterson (506), ion exchange is a common method for recovery of cadmium from industrial wastewaters, and many exchange resins are available with high specificity for the metal. To meet stringent effluent standards, some industries are using ion exchange rather than cheaper but less effective methods to treat chromate and chromic acid waters. With proper pH adjustment, chromate is removed even in the presence of high concentrations of sulfate and chloride (506). Reduction of hexavalent chromium to 0.023 mg/ℓ in a metal finishing wastewater has been reported (506). Cation exchange can be applied to remove trivalent chromium, and anion exchange can be employed to remove chromate and dichromate (506).

Ion exchange is capable of achieving very high levels of copper removal. Reduction of 1.02 mg/ℓ copper to less than 0.03 mg/ℓ has been reported. A selective ion exchange resin (Amberlite IR 120) reduced the copper concentration of an industrial copper plating rinse solution from 45 mg/ℓ to an undetectable amount (506). Koerts (359) found that ion exchange can remove copper and zinc from industrial waters to produce effluents containing as little as 0.04 mg/ℓ of copper and 0.1 mg/ℓ of zinc.

Removal of several metals, such as iron, manganese, lead, copper, and nickel can be accomplished with ion exchange, but the processes involve low pH or anaerobic water streams, which make them normally unsuitable for municipal water treatment. This technology, however, may be expected to develop as more attention is given to the application of ion exchange for the purification of reclaimed municipal wastewaters.

Ion exchange treatment of inorganic mercury-bearing waters appears to be capable of furnishing an effluent with 1 to 5 ppb inorganic mercury. Table 94 from Patterson (506) reviews the experience with ion exchange treatment for inorganic mercury. Effluent values in the ppb range are indicated. Preliminary tests have indicated that cation and anion exchange resins in series can remove 98 percent of both inorganic and organic mercury forms. Akzo Chemie has developed a special resin for mercury, which produces effluent levels below 5 ppb mercury (359).

TABLE 94. ION EXCHANGE TREATMENT
FOR INORGANIC MERCURY (506)

Resin Type	Treatment pH	Mercury (μg/ℓ) Initial	Mercury (μg/ℓ) Final	Additional Treatment	Percent Removal
Mtylon-T	5-6	5,000-25,000	1	--	100
Anion*	7	850	2.5	--	99.7
Macroreticular	na	10,000	<10	--	100
Anion*	na	470	30	--	94
Osaka IE*	"acidic"	3,000-10,000	100-150	Prefilter	98
Osaka MR*	na	100-150	2-5	Osaka IE Resin	98
Activex	na	10	<5	--	50
Ajinomoto	1.5	60	5	--	92
	6.0	87	3	--	97
	11.0	1,800	990	--	45
Billingsfors	6.5	35	1	--	97

*Mercury removed as mercuric chloride complex, $HgCl_x^{(2-x)}$.

Patterson (506) presented evidence that cation plus anion exchange applied to secondary wastewater effluent can remove selenium to a level of 99.7 percent. He also cited an example of 85 percent removal of silver from an extremely dilute secondary sewage effluent by cation exchange, and 91.7 percent removal by combined cation-anion exchange.

Calmon (100) discussed the use of ion-exchange for trace heavy metal removal. As part of his paper, he listed the specific resins that can be used to remove various heavy metals. These are as follows:

Element	Polar Group
Arsenic (3+)	Fluorone
Beryllium	Phosphonic diallyl phosphate
Bismuth	Pyrogallol
Boron	N-methyl glucamine, tris hydroxymethyl amino methane
Cesium	Phenolic OH + sulfonic groups
Cobalt	M-phenylene diamine, 8-hydroxyquinoline
Copper	Phenolic OH + phosphonic groups, 8-hydroxy quinoline m-phenylene diamine imino diacetic acid alginic acid
Germanium	Phorone
Gold	Pyridinium, thiourea

Element	Polar Group
Iron	Alginic acid m-phenylene diamine hydroxamic acid phosphonous phorone chlorophyll haemin deriv.
Lead	Pyragallol, phosphoric
Mercury	Thiourea, thiol, iminodiacetic acid, mercapto resins
Nickel	Alginic acid, dimethylglyoxime
Potassium	Dipicrylamine
Strontium	Phosphorous
Titanium	Chromotropic acid
Uranium	Pyridinium, phosphorous ester
Zinc	Anthranilic
Zirconium	Phosphate ester

He also discussed the various types of exchangers, dosages, operating problems, etc., that can be anticipated for removal of each metal.

BIOCIDAL CONTAMINANTS

Ion exchange units are not specifically designed to remove biocidal contaminants. However, some incidental removal of these pesticides, insecticides, and herbicides may occur due to adsorption on the resin material. This phenomenon will occur until the adsorptive capacity of the resin is exhausted.

SYNTHETIC/ORGANIC CONTAMINANTS

Because many of the synthetic/organic contaminants have chemical and physical properties that are similar to the biocidal contaminants, the same considerations apply here as in the biocidal section.

BIOLOGICAL CONTAMINANTS

Since the primary removal mechanism of ion exchange is based on particle charge properties, any removal of the biological contaminants will be incidental. Any removal that does occur will probably be due to mechanical filtration.

ADVANCED WATER TREATMENT: REVERSE OSMOSIS

INTRODUCTION

For readers unfamiliar with the osmotic membrane process, one can picture these membranes as thin sheet filter materials. Initial investigators thought that the membranes acted as strainer-type materials with such small pore sizes that most ionic and biological molecules were too large to pass through. Recent studies with electron microscopes have shown that removals are controlled by molecular diffusion through the membrane and that salts and other impurities diffuse much more slowly than water (652). The differential pressure to provide a driving force through the membrane is supplied by pumps. The discarded flow of concentrated contaminants is continuous, and its volume normally equals from 5 to 30 percent of the volume of the process influent volume. Membrane fouling by suspended solids, organic slimes, and precipitates is a problem unless substantial pretreatment of the influent water is provided. Therefore, reverse osmosis (RO) is normally located as the last unit process in the water treatment chain.

Very little actual operating data are available regarding the use of reverse osmosis for treatment of municipal water supplies, because the quality provided by reverse osmosis systems has not as yet been required. Even in areas of high TDS that could benefit from RO treatment, the cost of this unit process has been prohibitive. There has, however, been a substantial amount of research conducted on the use of RO for polishing highly treated wastewaters. This research, though, primarily focuses on the removal of selected contaminants from solution in laboratory and pilot scale projects. Since these are similar in many respects to raw water supplies, performance is often analogous to water treatment situations.

Reverse osmosis may become more important in the future as both a tertiary wastewater and raw water supply treatment process, because of its capability to remove a high percentage of all types of general, elemental, and biological contaminants as well as many synthetic/organic and biocidal constituents. With the recent concern over even very small concentrations of heavy metals, residual organics, and toxic compounds in water supplies, RO with its >99 percent removal efficiency may see increased usage, albeit at a high cost.

Aqueous solutions of two or more solutes reacting with a membrane may give product flux and solute retentions that are quite different from those predicted from the behavior of solutions of the individual solutes. This results from interactions between solutes, their products, and the membrane. Improved retentions of solutes have been observed when mixed in partially treated wastewaters, thus increasing the attractiveness of wastewater RO treatment for reuse (191). Improved retentions are thought to be due to synergistic effects among the high molecular weighted components.

Various types of membrane systems have been tested on a variety of wastewater effluents from primary to highly treated tertiary. Laboratory tests have also been conducted on the removal of many synthetic/organic chemicals and biocidal compounds. The RO process does not destroy any of the input contaminants, but only separates them into two streams, with the waste stream containing the rejected materials. Depending upon the feed conditions and the desired objectives, the product water volume can range up to 95 percent of the total influent, with the remainder representing the discarded flow. The characteristics of RO membranes can be controlled within a wide range by controlling the manufacturing variables. In general, as one improves the contaminant removal efficiencies, the flux per unit area decreases. This type of trade-off implies that the systems can be optimally designed to achieve the desired objectives.

For this study, the literature reviewed was oriented towards tertiary wastewater applications as well as laboratory and pilot scale tests of osmotic membranes. The literature excluded the extensive basic research on osmotic membranes for other applications (e.g., industrial wastewater treatment). The very extensive patent literature on the different types of membranes, designs, and manufacturing processes was also excluded. Table 95 provides a summary of the current literature pertaining to the performance of reverse osmosis units.

WATER QUALITY PARAMETERS

RO systems are not commonly used specifically for general contaminant removal from water supplies (with the exception of TDS), as these constituents can be sufficiently removed by other less expensive treatment units. However, there were quite a few references that noted the performance of RO on general contaminant removal from wastewaters as shown in Table 95. The performance of RO systems in terms of percent removal is excellent (90 to 99^+ percent) for all general contaminants except for low molecular components of BOD, or COD, and NO_3.

TABLE 95. LITERATURE REVIEWED PERTAINING TO REVERSE OSMOSIS

Contaminant	Reference Number
Water Quality Parameters	
ABS	46, 281, 286, 449, 593
NH_4	120, 148, 149, 182, 281, 294, 449, 593
BOD	148, 286
COD	120, 148, 149, 182, 189, 371, 449, 593, 615
Chlorides	120, 148, 149, 294, 449, 593, 671
CN	294, 671
Fluorides	148, 294, 671
Hardness	286, 294, 449
NO_3	140, 148, 182, 281, 286, 294, 593
PO_4	120, 148, 149, 182, 281, 286, 294, 449, 593
SO_4	120, 148, 149, 294, 449, 593
TDS	120, 148, 149, 182, 281, 286, 371, 449, 593
Elemental Contaminants	
Aluminum	148, 294
Arsenic	157
Barium	157
Boron	148, 157, 294, 671
Cadmium	157, 294, 615
Chromium	148, 157, 294, 615, 671
Copper	157, 294, 615, 671
Iron	157, 294, 449, 593, 615

TABLE 95 (continued)

Contaminant	Reference Number
Lead	615
Magnesium	120, 148, 149, 294, 449, 578, 593, 671
Mercury	157
Nickel	615, 671
Potassium	148, 149, 294, 449, 578, 593
Selenium	157
Silver	157, 615
Zinc	157, 615
Biocidal Contaminants	
DDT	46, 126
DDD	46, 126
Aldrin	126
Organophosphorus insecticide	126
Chlorinated hydrocarbons	126
Dieldrin	126
Herbicides	126
Lindane	46, 126
Pesticides	126, 191
Synthetic/Organic Contaminants	
Misc. organics	46, 148, 189, 281
Biological Contaminants	
Bacteria	148, 294
Virus	148

Several references discussed the removal of ABS from solution. The presence of highly nonbiodegradable alkyl benzene sulfonate (ABS) even in concentrations of only 1 mg/ℓ can produce undesirable frothing and foams although health implications do not appear significant. Hauck and Sourirajan (286) reported removals of 99^+ percent with a high feed concentration of 300 mg/ℓ. Merten and Bray (449) and an EPA study (593) both reported ABS rejection of about 98 percent with a variety of membranes. Bennett et al. (46) tested RO performance on a vast array of organics. They showed ABS removals of 90 percent at a 14 gpd/ft^2 flux.

Many studies were conducted that included ammonia removal (see Table 95). Reported removals ranged from 60 to 97 percent. Since ammonia concentrations are not generally of significance in water supplies and NH is readily oxidized in treatment processes, further discussion is not necessary here.

Hauck and Sourirajan (286) studied the performance of RO on hard water and on wastewater. The average BOD removal from secondary effluent was 85.8 and 80.8 percent at 1,000 and 500 psig, respectively. Cruver (148) reported secondary effluent BOD rejections varying from 81 to 94 percent with a cellulose acetate membrane.

COD removal is studied more frequently than BOD removal in regard to RO performance, because many of the nonbiodegradable constituents of COD can be effectively removed by RO. A review of all nine references reporting on COD removal shows a removal range of 90 to 99^+ percent with an average of about 95 percent. As a typical example, one of the larger scale pilot programs performed at Hemet and Pomona, California (615), found that RO provided very good removals of trace organics. The COD of secondary effluent was reduced from 39 mg/ℓ to 1 mg/ℓ at Pomona, while activated carbon effluent COD was reduced from 11.4 mg/ℓ to 0.3-1.0 mg/ℓ . Similar removals were reported from the RO pilot plant at Hemet. It was found that although RO was capable of greatly reducing effluent COD concentrations, the costs for full-scale operation are very high.

Chloride is readily removed by RO. Many studies have evaluated this constituent with removals ranging from 85 to 97 percent (120, 148, 149, 294, 449, 593, 671).

Two references included CN removal in research efforts (294, 671). Hindin and Bennett (294) found that CN removals ranged from 79 to 85 percent at a flux of about 18 gpd/ft^2. A summary report (671) listed typical CN removal at about 90 percent.

A study by Cruver (148) reviewed RO performance in removing various contaminants. It was found that fluoride removals ranged from 88 to 98 percent using selective cellulose acetate membranes. Hindin and Bennett (294) and reference 671 reported similar results.

RO units are highly efficient at removing hardness (Ca^{++} and Mn^{++}) from water supplies. Hauck and Sourirajan (286) summarized the effects of RO treatment of hard-water supplies in five cities and formal removals ranging from 96 to 99.9 percent as $CaCO_3$. Operating pressure was 1,000 psig with a 90 percent product recovery. Performance was directly related to flux, with the higher removals achieved at lower flow rates. Merten and Bray (449) also reported removals at roughly 99 percent for the more efficient membranes they analyzed.

The reviewed data indicates that nitrate nitrogen is one of the most difficult compounds for RO processes to remove (593). Hauck and Sourirajan (286) reported NO_3-N removals ranging from 50 to 60 percent. Extensive pilot testing by the EPA at Pomona, California, showed NO_3-N removals of 54 percent over the first 9,475 hours of operation (120). The five other references showed similar poor removals ranging from 51 to 86 percent and averaging around 60 percent.

Both $PO_4^{=}$ and $SO_4^{=}$, on the other hand, were very readily removed by RO processes. All references reviewed showed removals of 94 percent or better, some up to 99.9 percent.

The primary water treatment use of RO units is to reduce the TDS concentrations of highly mineralized waters. One full-scale example of this is the Orange County Water Districts Water Factory 21 (371) where 5 mgd of highly treated effluent is given RO treatment prior to recharge of potable aquifers. Results from the operation to date show a 90 to 95 percent TDS removal at 90 percent feed recovery. Results from other references using various membranes, pressures, and fluxes showed that average TDS removals were about 91 percent and ranged from 89 to 99 percent. Removals can be controlled by membrane selection and adjustment of flux. Studies of pilot RO systems at Pomona (120) showed TOC removals of 86 percent when using primary effluent on the feed.

The major problem areas associated with general contaminant removal are fouling, short membrane life, and associated engineering problems. If the membrane life and flux rate problems are solved, the RO membrane technology may become applicable to wastewater treatment where some sort of reuse is desirable and salt content would restrict reuse. At present, the tubular type membrane systems appear to be more applicable to sanitary wastewaters due to the lower fouling and easier cleaning inherent in this type of design. This advantage may not be as significant in potable water treatment.

ELEMENTAL CONTAMINANTS

A good deal of available literature on RO membranes deals with removal of elemental contaminants. RO systems can be designed to remove almost any elemental contaminant existing in either an ionic form or colloidal form in water. Generally, multivalent ions (Fe^{+3}, Cu^{++}, Zn^{++}, $SO_4^=$)are rejected more effectively than monovalent ions (B^+, NO_3^-). As previously mentioned, the percentage removal will depend upon the specific membrane and manufacturing procedures. Most references provide a lengthy list of contaminant removals. It would be redundant to discuss each of them here. References 154, 294, and 615 provide typical summaries of the performance of reverse osmosis units in removing elemental contaminants. Results are shown in Table 96. As shown, RO is generally very effective.

TABLE 96. REVERSE OSMOSIS REMOVAL OF ELEMENTAL CONTAMINANTS (REF. 154, 294, 615)

	Percent Removal (Single Pass)		
Contaminant	Reference 154	Reference 294	Reference 615
Aluminum	-	97	-
Arsenic	90-95	-	-
Barium	90-95	-	-
Boron	-	50	-
Cadmium	90-98	68-70	66-98
Chromium	90-97	93-98	82-98
Fluorides	90-97	88-98	-
Copper	90-97	82-96	99
Lead	90-99	-	99
Iron	90-99	95-98	94-99
Manganese	90-99	-	-
Mercury	90-97	-	-
Nickel	-	-	98-99
Selenium	90-97	-	-
Silver	90-97	-	96
Zinc	90-99	-	97

BIOCIDAL CONTAMINANTS

A few studies have been conducted on the removal of biocidals by osmotic membranes (46, 126, 191). Excellent removals were reported for a wide variety of pesticides, insecticides, and herbicides including chlorinated hydrocarbons, organophosphorus compounds, and halogenous cyclodienes. However, a considerable amount of this removal can be attributed to adsorption or absorption on the membrane itself. Since these tests were for short time periods relative to commercial application, the long-term rejection may be more complex and may depend upon whether the contaminant is adsorped or absorbed and upon the diffusion rates through the membrane.

With such a large percentage of the removal being related to adsorption/absorption, it is clear that different types of membrane materials will show significantly different results. For example, cellulose acetate (CA) membranes show rather poor performance on the more polar randox and atrazine, whereas the less polar polythylenimine-based membranes showed satisfactory performance. Since the active surface on some types of membranes is on the order of a few tenths of a micron, any significant amount of absorption can change the membrane composition and possible rejection of other contaminants.

Edward and Schubert (191) summarized some results of various studies using cellulose acetate (CA) membranes for the removal of the common pesticide 2, 4-D. Results varied widely from 57 to 99+ percent removal depending on influent concentrations, esterification, and feed rate. Performance for removal of the insecticide lindane was also sporadic, ranging from rather poor removals up to 84 percent. DDT and DDD have a very low solubility and tend toward micelle formation thus enhancing RO performance. Reported DDT and DDD removals were 97 to 99 percent (157, 191).

Chian et al. (126) performed a detailed analysis of the performance of RO units in removing biocidals. Two types of membranes were evaluated, cellulose acetate (CA) and cross-linked polyethylenimine (NS-100). With each membrane, rejection of all types of pesticides (chlorinated hydrocarbons, organophosphorus, and miscellaneous) was greater than 99 percent. Specifically, the following chlorinated pesticides were retained at greater than 99 percent: aldrin, lindane, heptachlor, heptachlor epoxide, DDE, DDT, and dieldrin.

Of the organophosphorus pesticides, diazinon was removed at greater than 98 percent, and methylparathion, malathion, and parathion at greater than 99 percent. The lowest removals were reported with the CA membrane on randox (72 percent) and atrazine (84 percent). A significant portion of the pesticide removed was adsorbed on the membrane itself: about 80 to 95 percent for hydrocarbons, 30 to 50 percent for organophosphorus compounds, and 5 to 60 percent for the miscellaneous pesticides.

Bennett et al. (46) reported on the removal of organic refractories including some biocidals. They found that lindane removal was 84 percent; DDT, 99^+percent; and casein, 99 percent.

The literature data indicate that removal of biocidals is highly variable, depending not only on contaminant concentrations and membrane characteristics, but on synergistic effects of other components in the water. In any case, RO provides a high level of treatment for many of the common pesticides and insecticides.

SYNTHETIC/ORGANIC CONTAMINANTS

The performance of RO membranes with respect to removal of synthetic/organic contaminants is similar to that of many of the biocides. In general, these contaminants can be adsorbed, absorbed, rejected or transmitted by the membrane, with removals depending on chemical species and membrane type. In general, larger molecular weight compounds are readily rejected or sorbed by the membrane, whereas low molecular weight compounds (<100) are more likely to pass through.

Studies by Duvel and Helfgott (189) on cellulose acetate (CA) type membranes have shown that rejection of low molecular wet organics depends upon the molecular weight and molecular size (as determined by steric geometry, with the size being more important); it also depends on the ability of the molecule to form hydrogen bonds. The hydrogen bonding behavior affects the solubility in the membrane surface and hence the permeability.

Studies by Hamoda et al. (281) have shown that high flux membranes can be developed that have good rejections (>99 percent) of tested organic compounds such as sucrose, glutamic acid, starch, sodium stearate, ABS, LAS, and beef extract. Cruver (148) reviewed similar contaminant removals and listed sucrose at 99.9 percent and glucose at 99.5 percent.

Bennett et al. (46) performed a comprehensive study of the removal of organic refractories by RO. Results showed that the RO process is capable of producing a product water that is extremely low in organic matter in aqueous solution or dispersion, with the exception of those organic compounds that, when in solution or dispersion, have a lower vapor pressure than water.

The classes of compounds that do not appear to be well rejected and are present in wastewater effluents include compounds such as methanol, ethanol, and phenol. No data were found on the low-molecular-weight, halogenated hydrocarbons such as chloroform, bromoform, halogenated ethane, or ethylene compounds; but, based on the present theoretical knowledge, one would expect the removals to be poor.

BIOLOGICAL CONTAMINANTS

Due to the large size of biological contaminants, including virus, relative to the effective pore size of RO membranes. high reductions of these contaminants can be expected.

Hindin and Bennet (294) conducted microbiological studies to determine the permeation through a porous cellulose acetate membrane of microorganisms found in sewage effluent. Their results showed that *Escherichia coli*, *Aerobacter aerogenes*, coliphage T-7 and X-175, and *Streptococcus narcesence* were all removed 100 percent by the RO unit, with the exception of one test in which a leak in the membrane may have permitted permeation.

Cruver (148) reports that several studies have shown that 99.9 percent removals of bacteria and virus can be attained. However, even with these excellent removals, RO processes are not used alone for disinfection because of the presence of imperfections in the membranes. These systems are primarily designed for TDS removal in which small leaks through the membrane and at seal joints are generally inconsequential. Nevertheless, these leaks could be significant when they reduce the removal of virus and bacteria from 99.9999 to 98 percent. To depend upon these systems for 100 percent biological contaminant removal would require continuous monitoring for biologicals and a degree of quality control that would be considered beyond the state of the art for field systems.

The present literature indicates that whereas removals of biological contaminants with RO are very good, they are not as high or as fail safe as other disinfection practices (chlorination, ozonation). It should also be remembered that RO is only a separation process, not a destruction unit, and that the biological contaminants, once removed, will remain in the waste solution.

SECTION 11

EPIDEMIOLOGICAL AND PATHOLOGICAL EVALUATION OF WASTEWATER CONTAMINANTS

INTRODUCTION

Many contaminants contained in municipal wastewater are known to produce adverse human health effects. A number of elemental, biocidal, and synthetic/organic constituents have been clearly identified as potential carcinogens; some have even been identified as being mutagenic or teratogenic. Although the epidemiological effects of various viruses and bacteria are known and predictable, knowledge of the health risks of certain other contaminants is incomplete or lacking. Moreover, some chemical substances work not only in isolation within the human body, but may react synergistically (two or more chemicals combining to produce a net effect that is greater or lesser than that produced if the chemicals act independently). A multiplicity of factors is involved in such reactions, and knowledge of potential health risks is scant.

Literature detailing the epidemiological and pathological effects of wastewater constituents has been surveyed; the principal health problems posed by these contaminants are summarized below. A sound understanding of human physiological reactions and consequent public health threats will better prepare responsible authorities to assess treatment performance and set acceptable standards.

Present standards controlling wastewater discharges and drinking water supplies have been assumed by many to guarantee adequate protection of public health. However, recent investigation has shown that some residual organics, carcinogenic chlorinated hydrocarbons, synthetic compounds, trace elements, and biocides are harmful even in extremely small concentrations. Existing standards are called into question by this increasing knowledge: there may be no "safe" threshold for some of these chemicals. Growing epidemiological and pathological evidence must be taken into account if discharge and drinking water standards are to safely ensure public well-being.

WATER QUALITY PARAMETERS

Suspended solids, BOD, TOC, and most other constituents of general water quality have no direct effect on public health. However, certain constituents may become associated with other, more directly harmful, contaminants. The survival and potential health threat of such contaminants may be magnified as a result of this process. Moreover, nitrogen species present in wastewater can directly affect human health.

Nitrogen

Nitrogen in wastewater effluent is usually found in one of the stable forms (ammonia, nitrate, or organic nitrogen) rather than the more hazardous nitrite form. Nitrates and nitrites occur in drugs, food, and water. Man is continually exposed to small amounts of these substances, which usually cause no harm. In high concentrations and under special circumstances, however, they may cause illness and even death. Sapp (572) and the Hazardous Waste Advisory Committee of the EPA (484) consider nitrite, in particular, a significant health problem.

Nitrite toxicity is the major health problem associated with these nitrogen species since nitrate easily reduces to the toxic nitrite form. Such a conversion may occur outside the human body in food or water containing nitrates. Conversion can also take place inside the body through the action of intestinal bacteria on ingested nitrates.

Nitrate/nitrite conversion that occurs during digestion requires special conditions likely to be present only in infants. The foremost prerequisite is the presence of nitrate-reducing bacteria in the upper gastrointestinal tract. Such bacteria are not normally present so high up in the intestinal tract. This circumstance, however, may occasionally occur in infants, particularly those with gastrointestinal infections and a gastric pH insufficiently acidic to kill the bacteria.

Acute nitrite toxicity (methoglobinemia) occurs when hemoglobin (the oxygen-carrying red pigment of blood) is oxidized into methemoglobin (a brown pigment incapable of carrying oxygen). Methemoglobin constitutes about 1 percent of the total hemoglobin of a healthy adult and up to approximately 4 percent of that of a healthy newborn infant. Cyanosis results when roughly 15 percent of the hemoglobin in blood is converted into methemoglobin; when methemoglobin constitutes 70 percent or more of the total hemoglobin, oxygen transport is severely impeded, and death may occur (484).

Infants, then, are particularly prone to nitrate-induced methemoglobinemia. In addition to the presence of nitrite-reducing bacteria in the upper gastrointestinal tract as mentioned above, (1) the hemoglobin of very young infants is oxidized twice as rapidly as that of adults by nitrate to form methemoglobin; (2) the red blood cells of infants are not able to reduce methemoglobin into hemoglobin as well as adult cells; and (3) the total fluid intake of infants per unit of body weight is much greater than that of adults. Thus, for a given concentration in fluids, infants consume proportionately more nitrate than adults.

The consumption of water containing high levels of nitrates has accounted for many more cases of methemoglobinemia than all other causes combined. Methemoglobinemia of such etiology has been reported only in infants, although one study documents one occurrence resulting from the use of nitrate-contaminated well water for peritoneal dialysis in an adult. In the United States only one case has been associated with water from a public water supply; all the rest (about 300) have been due to well water (484).

Standards for nitrates in drinking water limiting nitrate to 10 mg/ℓ expressed as nitrate-nitrogen (45 mg/ℓ expressed as nitrate) were set by the U.S. Public Health Service in 1962. The 10-mg/ℓ nitrate-nitrogen level was set because there had been no reports in this country of infantile methemoglobinemia associated with the ingestion of water containing nitrate at levels below 10 mg/ℓ. In addition, this level was set because it was a standard that could be met easily by most municipal water supplies. After publication of these standards, however, data reported from other countries revealed that a small percentage of cases had occurred where the water nitrate-nitrogen content had been below 10 mg/ℓ.

As a consequence, the 1962 standards are currently under reevaluation. Several studies have been designed to determine more specifically the exact nitrate levels in water required to cause elevated levels of methemoglobin and clinical evidence of methemoglobinemia in infants. Preliminary results suggest that the 1962 standards provide adequate protection against clinical methemoglobinemia. However, subclinical elevations of methemoglobin have been found in infants with diarrhea or respiratory disease, consuming water with a nitrate content below the 10 mg/ℓ level (484).

In contrast to the relative wealth of data on acute toxicity in humans, reliable data are lacking on the physiologic effects, if any, of chronic nitrate/nitrite toxicity or of mild, noncyanotic methemoglobinemia. Studies in animals indicate that nitrates and nitrites may, on occasion, cause vitamin A deficiency, and that nitrates may have an antithyroid effect by increasing the need for iodine. No data are available to indicate whether such effects can occur in man.

Chronically elevated levels of methemoglobinemia may have some effect on the human brain: abnormal changes on electroencephalograms have been observed in rats given 100 to 2,000 mg/ℓ of sodium nitrite each day for two weeks. A Russian study purports to show decreased response to visual and auditory stimuli in school children with a mean methemoglobin level of 5.3 percent of total hemoglobin. However, the study was poorly controlled, and the results are inconclusive. There have been patients with hereditary methemoglobinemia and mental retardation, but the association may be coincidental. Most patients with hereditary methemoglobinemia show no mental or neurologic abnormalities (484)

Nitrosamines formed by the reaction between nitrites and amines have been proven hazardous to human health. Nitrites and/or precursor nitrates are found in foods, water, and drugs; amines are found in tobacco smoke, beer, tea, wine, and toothpaste as well.

Nitrosamines have potent biological effects, including acute cellular injury (primarily involving the liver), carcinogenesis, mutagenesis, and teratogenesis. To date, approximately one hundred nitrosamines have been tested in animals. The vast majority has proved to be carcinogenic. Many organs (liver, esophagus, and kidneys) that are common to diverse species of animals are susceptible to the cancer-producing effects of these compounds. These effects can be elicited experimentally by various routes of nitrosamine administration (oral, intravenous, inhalation) at extremely low doses. In some instances, cancer can develop after a single exposure (484).

Concerns about potential nitrosamine hazards to human health arise from the possibility for (1) contact with preformed carcinogenic nitrosamines and (2) the formation of carcinogenic nitrosamines within the human body after exposure to precursor nitrites and amines. The possible formation of carcinogenic nitrosamines in the human gut through the combination of ingested nitrites and amines is of critical concern. Such reactions have been demonstrated to occur both in vitro and in vivo (in animals).

Studies in humans that were fed nitrate and a noncarcinogenic nitrosamine precursor amine (diphenylamine) have shown that diphenylnitrosamine can be formed in the human stomach. Nitrosamine determinations in these studies were made by thin-layer chromatography, a method now known to give false positive results. Unfortunately, these data have not yet been confirmed using the newer and more reliable techniques of gas-liquid chromatography and mass spectrophotometry (484).

ELEMENTAL CONTAMINANTS

It is often difficult to assess the health effects of metals and their compounds: many metals are essential to life at low concentrations but toxic when concentrations exceed tolerance in man. The situation is further complicated because the various chemical states of metals (pure metal, inorganic or organic-metallic compounds) react differently within the body. Individual differences between subjects, incubation periods, and sites of accumulation of the substances in the body are also significant factors in toxicity.

In addition, results from experiments with animals may not be readily applicable to humans. Experimental animals such as rats and mice have a much shorter life-span than man and react and respond differently to chemicals because of their own distinctive physiological processes.

Table 97 gives a comprehensive summary of the presence of metals in the environment, their toxicity to humans, and their half-life in the body (the time it takes for one-half the chemical to be excreted). Five heavy metals -- cadmium, lead, mercury,nickel as nickel carbonyl, and beryllium -- represent known hazards to human health. Lead, mercury, and cadmium are particularly insidious,because they can be retained in the body for a relatively long time and can accumulate as poisons. Antimony, arsenic, cadmium, lead, and mercuric salts are the most toxic. Discussions of fatal doses and other considerations can be found in each individual metal section.

Lead

Lead is a cumulative poison. However, except in cases of prolonged exposure at high concentrations, most of it is absorbed into the blood and is later excreted in the urine. The blood lead does not rise to acute levels; however, a small portion of the daily lead intake gradually accumulates in bones, where it is normally insoluble and harmless. Under certain conditions, such as periods of high calcium metabolism in feverish illness, cortisone therapy, or old age, this accumulated lead can be released suddenly into the blood at toxic levels. A fatal dose of absorbed lead has been estimated to be 0.5 g; ingestion of more than 0.5 mg/ℓ/day may, because of the above-mentioned accumulation, cause toxicity and death.

TABLE 97. METALS IN THE ENVIRONMENT AND THEIR TOXICITY[a]

Metal	(ppb) Food and Water	(ppb) Air	Oral dose producing toxicity (mg)	Fatal dose (ingestion)	Total Body content (mg)	Whole body half-life (days)
Antimony	100	1.7	100	100-200 mg	7.9	38
Arsenic	400-900	--	5-50	120 mg	15-20	280
Barium	735	30	200	1 g	22	65
Beryllium	12	0.04	--	--	0.3	180
Cadmium	20 to 100	7.4	3	--	50	25 years
Chromium	245	1.1	200	5 g	1.8	616
Copper	1,325	11.4	50-250	10 g[b]	72	80
Iron	15,000	84	--	5-10 g[c]	4,200	800
Lead	300	46	--	0.5 g	120	1,460
Manganese	4,400	28.8	--	--	12	17
Mercury	25	--	--	20 mg - 1 g[d]	--	70[e]
Nickel	600	2.36	--	--	10	667
Selenium	62	--	5	--	14.6	11
Silver	60-80	--	60	2 g[f]	1	5
Tin	7,300	0.6	2,000	--	17	35
Zinc	14,500	16.8	--	10 g[g]	2,300	933

[a] Data primarily from Dreisbach (181) and Oregon State University (288)

[b] Copper sulfate

[c] A two-yr old child

[d] Mercuric salts

[e] Methyl mercury

[f] Silver nitrate

[g] Zinc sulfate

Lead prevents the formation of hemoglobin in the blood by interfering with the synthesis of certain precursors (prophyrins), which leads to the anemia present in chronic lead poisoning. Lead also inhibits the sulfhydryl enzymes that catalyze many of the metabolic pathways, including the biosynthesis of heme. The effects of lead on the brain and peripheral nervous system are most serious, manifested as gastrointestinal or central nervous system disturbances and anemia. Symptoms of acute poisoning include metallic taste, abdominal pain, vomiting, diarrhea, black stools, oliguria. collapse, and coma. Symptoms of chronic poisoning in early stages include loss of appetite and weight, fatigue, headache, lead line on gums, loss of recently developed skills, and anemia. In advanced stages, there is intermittent vomiting, irritability; nervousness; lack of coordination; vague pains in arms, legs, joints, and abdomen; paralysis; disturbances of menstrual cycle; and abortion. Exposure to tetraethyl lead or tetramethyl lead causes insomnia, disturbing dreams, emotional instability, hyperactivity, and even toxic psychosis. Severe symptoms include persistent vomiting, papilledema, ataxia, encephalopathy (any disease of the brain), elevated blood pressure, cranial nerve paralysis, delirium, convulsions, and coma (131).

There are up to 100 cases of lead poisoning reported in the U.S. annually; an average of 10 are fatal. Most of the fatalities are related to children who ingested lead-based paint from homes built before 1940; however, 7 cases of lead poisoning were reported from drinking well water in Australia in 1973 (57, 144). The well water contained a soluble lead content of about 14 mg/ℓ.

Mercury

Neither methyl nor elemental mercury is normally found in dangerous concentrations in air, water, or most common foodstuffs. There are three principal ways in which man can be poisoned by methyl mercury: (1) when food is consumed that has been contaminated with methyl mercury, e.g., seed containing mercury fungicides ($HgCl_2$); (2) when methyl mercury used or formed in industrial processes is intentionally or unintentionally dumped into natural waters, reaching man directly through the water and indirectly through the food chain; and (3) when nontoxic inorganic or organic phenyl mercury is converted into toxic alkyl mercury compounds by microorganisms in the environment and is passed on to man through the food chain (329).

The acute toxicity of methyl mercury is the result of almost complete (98 percent) absorption of the compound from the gastrointestinal tract. Ingested metallic mercury is not toxic since it is not absorbed. Mercurous chloride and organic mercurials such as acetomeroctol, ammoniated mercury, merbromin, mercocresol, and mercury protoiodide are not likely to cause acute poisoning because they also are poorly absorbed. The single fatal dose of these compounds is 3 to 5 times the fatal dose of soluble mercury salts. The mercurial diuretics (mersalyl, meralluvide, mercurophylline, mercumatilin, mercaptomerin, chlormerodrin, and merethoxylline) are almost as toxic as mercury salts. Volatile diethyl and dimethyl mercury are 10 times as toxic as mercuric chloride (695). The fatal dose of mercuric salts is 20 mg to 1 g. The biological half-life of methyl mercury is estimated to be about 70 (30 to 100) days as shown in Table 97 (615,672).

Acute poisoning by ingestion of mercuric salts causes metallic taste, thirst, severe abdominal pain, vomiting, and bloody diarrhea. Death is from uremia (an excess of urea and other nitrogenous waste in blood). Ingestion of insoluble or poorly dissociated mercuric salts (including mercurous chloride and organic mercurial compounds) over a prolonged period causes urticaria progressing to weeping dermatitis, stomatitis, salivation, diarrhea, anemia, liver damage, and renal damage progressing to acute renal failure with anuria (total suppression of urine). In children, repeated administration of calomel (Hg_2Cl_2) appears to be the cause of a syndrome known as erythredema polyneuropathy. In fatalities from mercury poisoning, the pathologic findings are acute tubular and glomerular degeneration or hemorrhagic glomerular nephritis. The mucosa of the gastrointestinal tract shows inflamation, congestion, coagulation, and corrosion (181).

Mercurialism is manifested primarily in kidney, liver, or brain damage in animals. Exposure to inorganic mercury compounds usually results in kidney damage, while alkyl mercurialism is characterized by brain damage. However, some degree of both kidney and neurological injury results from exposure to either category of mercurials. Mercury poisoning apparently damages Kreb's cycle enzymes (which catalyze the oxidation of tricarboxylic acids) and protein synthesis, leading to kidney and brain damage (216).

Methyl mercury has an affinity for the fetus and is teratogenic in its effect, as it readily penetrates to the fetus through the placenta. In addition, the cytogenic toxicity of methyl mercury is potentially greater than that of any other known substance. The urine and especially the feces are the most important means of mercury elimination.

The tragedy of Minamata Bay, Japan, which occurred from 1953 to 1961, is one of the best documented cases of mercury poisoning. In essence, it was concluded that the disease -- which had felled many inhabitants of the fishing villages on the shores of Minamata Bay -- had resulted from mercury poisoning. A chemical plant in the area used mercury chloride as a catalyst in the production of vinyl chloride. The waste containing the mercury washed off the product was discharged into the bay, and was ingested by the shellfish therein. Some 114 cases of "Minamata disease" were reported, as well as 44 deaths and 22 cases of brain damage in 400 live births (423).

A similar disaster struck at Niigata, Japan, where 120 persons were poisoned (423).

Nickel

The fatal dose of nickel is not know, but its whole body half-life is about 667 days. Inhaled nickel carbonyl decomposes to metallic nickel, which deposits on the epithelium of the lung. This finely divided nickel is rapidly adsorbed and damages the lung and brain. The principal manifestation of nickel carbonyl poisoning is dyspnea (difficult respiration). Workers exposed to nickel carbonyl show a high incidence of lung cancer, and some workers develop dermatitis (181). Very little is understood about the adverse health effects of nickel in waste or water supplies.

Cadmium

Cadmium has become the most recent and perhaps the most acute menace among the widely used heavy metals. A great amount of current research is being conducted regarding the fate and distribution of cadmium to the environment.

Nearly all this literature concerns the quantification of cadmium in wastewater and sludges, and the effects of disposal to land and water systems. However, there is little information available concerning the direct health hazards of cadmium present in wastewater and water supplies. The average American citizen's daily intake of cadmium from foods and water supplies is estimated to be between 0.02 and 0.1 mg/d. The oral dose of cadmium producing toxicity is about 3 mg, but its fatal dose is not known. The whole body half-life of cadmium is 25 years (Table 97).

Circumstantial evidence appeared to point to some link between trace metals of cadmium and hypertension. Recent studies, however (615), have disagreed with this finding, and the general consensus now is that there is no link between cadmium ingestion and hypertension. Inhalation of tobacco smoke is a major source of cadmium accumulation in man. Only about 5 percent of the cadmium ingested through food or drink is absorbed by the body, while 10 percent to perhaps as much as 50 percent of inhaled cadmium is retained (615).

Cadmium tends to accumulate in liver and kidney tissues because of its very long biological half-life in man (estimates range from 10 to 25 yr, compared with about 70 days for methyl mercury). Excessive levels in the kidney cortex (over 200 mg/g wet weight) results in proteinuria. Therefore, the cadmium concentration in water must be kept low (615). The Environmental Protection Agency (EPA) in its 1975 Interim Primary Drinking Water Standards set a mandatory limit of 0.010 mg/ℓ for cadmium concentrations in drinking water; the World Health Organization set a limit of 0.05 mg/ℓ . The results of a U.S. Geological Survey investigation of 720 waterways showed that 4 percent had concentrations above EPA standards (423).

Cadmium has reportedly caused a number of deaths from oral ingestion of the metal in food or water. For example, Japanese people living along the Jintsu River suffered for years from an unknown malady characterized by kidney malfunction, a drop in the phosphate level of the blood serum, loss of minerals from the bones, and osteomalcia resulting in bone fractures causing intense pain. One of several causes of the malady implicated was a cadmium, zinc, and lead mine that was discharging wastewater into the river. The disease, known as itai itai, was contracted either by drinking water from the river or by eating rice that had accumulated the metal from irrigation water (123). In another situation, there was an outbreak of acute gastroenteritis in 13 children who drank orange soda contaminated with cadmium at a concentration of 16 mg/ℓ . The contamination was caused by the orange soda coming in contact with the soldered joints in the tank of the soft drink machine (57, 60).

Chromium

Chromium, which exists in various oxidation states (+2+3 and +6), appears to be most toxic to man as the hexavalent chromium ion. The fatal dose of a soluble chromate such as potassium chromate, potassium bichromate, or chromic acid is approximately 5 g. Acute poisoning from ingestion is manifested by dizziness, intense thirst, abdominal pain, vomiting, shock, oliguria (scanty urination), or anuria. Hemorraghic nephritis occurs, and death is from uremia.

Repeated skin contact with chromium leads to incapacitating eczematous dermatitis with edema and slowly healing ulceration. Breathing chromium fumes over long periods of time causes painless ulceration, bleeding, and perforation of the nasal septum accompanied by a foul nasal discharge (181).

Whether chromium is carcinogenic is questionable at this time. However, the incidence of lung cancer in workers exposed to dusty chromite, chromic oxide, and chromium ores is reported to be up to 15 times the normal rate (181).

Experiments on rats showed no toxic response from drinking water containing 0.45 to 25 mg/ℓ in chromate and chromium ion form (408).

Although a potential health hazard exists, evidence of health problems resulting from chromium present in wastewaters is lacking in the literature.

Arsenic

Arsenic is widely distributed in nature. It is present in toxic concentrations in many water supplies: cattle in New Zealand have died from drinking water containing natural arsenic, and there are several areas of the world where there is a high incidence of skin cancer among people drinking well water that contains natural arsenic (423). In 1971 the U.S. Geological Survey found that 2 percent of the samples drawn from 720 waterways were above their standard of 0.05 ppm for arsenic.

Before the advent of modern insecticides, arsenic compounds were widely used to treat food crops. Although arsenic can stimulate plant growth in very low concentrations, in excessive quantities, as little as 1 ppm of arsenic trioxide arsenic can be injurious. Organic arsenicals, such as arsphenamine and dimethylarsinic acid, release arsenic slowly and are less likely to cause acute poisoning. But arsenic accumulates in the body, so decreasingly small doses can be lethal. Repeated or prolonged intake has a cumulative toxic effect, presumably caused by the arsenic combining with sulfhydryl (-SH) enzymes and interfering with cellular metabolism.

Chronic poisoning from ingestion or inhalation of arsenic can cause anemia, weight loss, polyneuritis, optic neuritis, dermatitis, cirrhosis of the liver, abdominal cramps, chronic nephritis, and cardiac failure (181).

Arsenic is suspected to be carcinogenic but not tumorigenic (29, 340, 363). Chronic exposure to arsenic-contaminated water of 0.3 mg/ℓ is also suspected to be related to the increased incidence of hyperkeratosis (hypertrophy of the horny layer of the epidermis) and skin cancer; chronic exposure at levels of 0.8 mg/ℓ may be related to gangrene of the lower limbs (265).

Arsenic is one of the impurities in mineral phosphate deposits, a source of commercial water softeners. Concentrations of 10 to 70 ppm of arsenic have been detected in several common household detergents. Baby rash, hand rash, skin eruptions, and other types of dermatitis allergies are associated with arsenic in detergents. Much of the sewage containing such detergents is dumped into waterways (423). There is a danger that arsenic in laundry water may be absorbed through unbroken skin.

The principal oxidation states of arsenic are +3 and +5. In the +3 state, arsenic forms arsenious oxide; in the +5 state, arsenious acid; and in the -3 state, arsine gas. The fatal dose of arsenic trioxide is about 20 mg. Arsenic poisoning is manifested by gastrointestinal disturbances; death is due to circulatory failure as a result of hemolysis (destruction of the red blood cells).

Copper

The biological properties of copper are such that it is a useful biocide, especially as an algicide. Various salts of copper (one of the most common being copper sulfate - $CuSO_4$) are used as astringents, deodorants, and antiseptics. These salts are all water soluble, and their protein-precipitating characteristics form the basis of their astringent and antiseptic effects. The copper mined in the United States for these effects alone amounts to some 15 million lb/yr and accounts for about 40 percent of all chemical uses of the metal.

Trace amounts of copper are essential for normal metabolism, and relatively large concentrations can be tolerated by most animals including vertebrates. The effect of copper on aquatic organisms varies greatly; microorganisms, including algae, are highly susceptible.

Small quantities of copper are not considered toxic, but higher concentrations are known to cause vomiting and liver damage (672). Fatalities have been reported following the ingestion of 10 g of zinc or copper sulfate (181). An outbreak of acute copper poisoning occurred in a school in Mesa, Arizona. The outbreak began 10 min after the students drank an orange-flavored drink that had been kept in a brass container for 17 hr (57). An 8-oz glass of the drink contained 8.5 mg of copper.

Selenium

Selenium is an excellent example of the fine balance that can occur in nature between the beneficial and injurious effects of a natural substance. An essential micronutrient for some plants, selenium is one of the most toxic substances to occur naturally in the environment; concentrations only slightly above those needed for growth of plants may be poisonous to animals. A selenium compound that was used to kill insect pests in fruit orchards left a residue in the fruit. Cattle that were feeding on this fruit contracted a chronic form of livestock poisoning called alkali disease. Ingestion of forage containing about 25 ppm can cause this disease when the forage is eaten for several weeks or months (423).

As long ago as 1936, inhabitants of seleniferous areas of South Dakota and Nebraska complained of gastrointestinal symptoms and were found to excrete large quantities of selenium in their urine (423). There is also a report of selenium toxicity from a three-month exposure to well water containing 9 mg/ℓ of selenium.

The American Conference of Governmental Industrial Hygienists recommends a selenium limit of 0.2 mg/cu m in air and 0.01 ppm in water.

Beryllium

Although the fatal dosage of beryllium is not known, in 1971 the EPA placed beryllium on the list of hazardous pollutants. From 1941 to 1967, 760 cases of berylliosis were recorded (181).

Soluble beryllium salts are irritating to the skin and mucous membranes, and induce acute pneumonitis with pulmonary edema. At least part of the changes present in acute pneumonitis and berylliosis (chronic pulmonary granulomatosis) develop from hypersensitivity to beryllium in the tissues. Weight loss and marked dyspnea, which are symptoms of berylliosis, begin 3 months to 11 years after the first exposure (inhalation). Eczematous dermatitis with a maculopopular, erythematous visicular rash appears in a large percentage of workers exposed to beryllium dusts.

Barium

Absorbable salts of barium, such as the carbonate, hydroxide, or chloride, are used in pesticides; the sulfide is sometimes used in depilatories for external application. A soluble barium salt such as the carbonate or hydroxide may be present as a contaminant in the insoluble barium sulfate used as a radiopaque contrast medium.

The fatal dose of absorbed barium is approximately 1 g. The principal manifestations of barium poisoning are tremors and convulsions. Barium presumably induces a change in permeability or polarization of the cellular membrane that results in stimulation of all cells indiscriminantly (423).

Other Elements

Antimony will be considered a potential hazard if levels of the element increase. It is present chiefly in industrial wastes, typesetting metal, pewter, and enamel ware. Antimony is suspected to cause a shortened life-span and heart disease in rats (423).

Metal alloys and smoke suppresant in power plants are the major sources of manganese as a pollutant. Although manganese in trace amounts is an essential element to man, increased levels may imperil health. The findings in one death - suspected to be caused by the ingestion of manganese-contaminated drinking water - were atrophy (a wasting of the tissues) and disappearances of cells of the globus pallidus (in the brain). Experimental animals show inflammatory changes in both gray and white matter (423).

BIOCIDAL CONTAMINANTS

There is significant information in the literature concerning biocidal contaminants, their chemical and physical characteristics, toxicology, analytical chemistry, and impacts on health and the environment. In both chemical and medical literature, hundreds of cases of acute poisoning resulting directly or indirectly from such pesticides have been reported. Table 98 summarizes the various levels at which LD_{50}, fatal dose, chronic poisoning, and acute poisoning occur.

Chlorinated Hydrocarbons

Because of their persistence and nondegradational characteristics, many of the relatively less toxic chlorinated hydrocarbons, such as DDT, aldrin, and dieldrin, have been banned and are being replaced by highly toxic but less persistent organophosphorus pesticides. The U.S. EPA is presently conducting a program to identify new, less harmful pesticides that can act as substitutes for those causing the problems.

This decision to remove aldrin and dieldrin from the market has been highly criticized. The idea that these compounds pose "an unreasonable risk of cancer" is based on the presence of tumors in the livers of mice that were given food containing aldrin and dieldrin; some of these tumors metastasized to the lungs. However, it has been noted that similar tumors can be produced by other compounds, such as DDT and phenobarbital, and can occur in mice on normal diets. There is, then, some question as to whether the production of tumors in the livers of mice given aldrin or dieldrin is a reliable indicator of a hazard to man. The general dietary intake of all major pesticides except aldrin and dieldrin

TABLE 98 . BIOCIDES IN THE ENVIRONMENT AND THEIR TOXICITY*

Biocide	LD_{50} (oral) mg/kg	Fatal Doses (g/kg)	Chronic Poisoning	Acute Poisoning
DDT	Rat 285 Rabbit 325	0.4	Not substantiated. Having 648 ppm in their body remained well.	Severe vomiting within 30 min to 1 hr of 5 g. Weakness and numbness of the extremities. Apprehension and excitement are marked.
Dieldrin	Rat 60 Dog 68 Rabbit 45	0.07	Not been established in man. Impair liver function in animals, occasional epileptiform convulsions.	Hyperexcitability, tremors, ataxia, convulsions.
Lindane (Benzene hexachloride)	Rat 135 Dog 120 Rabbit 130	0.6	In animals, liver necrosis.	Vomiting and diarrhea, convulsions, circulatory failure.
Malathion	Rat 2500	0.86	In animals, colinesterase levels of red blood cells and plasma are reduced markedly.	Headache, tremors, nausea, abdominal cramps, diarrhea, coma, heat block.
Parthione	Rat 4	0.0014	Not established in man.	Similar to those of malathione, but more severe and fatal.

TABLE 98 . (continued)

Biocide	LD_{50} (oral) mg/kg	Fatal Doses (g/kg)	Chronic Poisoning	Acute Poisoning
2, 4-D	Mouse 375	0.7	Weakness, fall of blood pressure, muscle damage.	Burning pain, painful and tender muscle, fever, paralysis, irreversible fall of blood pressure.
2, 4, 5-T	Rat 300 Dog 100	0.6	Similar to 2, 4-D	Similar to 2, 4-D.

* Data primarily from Sunshine (626), Dreisbach (181), and McKee and Wolfe (436).

is well below the allowable standards that were established in 1972 by the World Health Organization Council on Environmental Quality. Much of the DDT uptake (about 85 to 90 percent) comes from food; the remainder comes from air, water, aerosols, cosmetics, and clothing.

Because of DDT's extremely low solubility in water (about 2 ppb), the body, which is essentially a water system, cannot handle the liquid soluble substance and deposits it in fat. Quantities of DDT and other related pesticides do not appear to build up continuously; instead, the pesticides reach a plateau or steady state (storage equilibrium) at which they are being excreted and degraded at levels equal to their intake. Children may take 5 to 10 yr to reach storage equilibrium. Little is really known about these levels, which undoubtedly vary with different pesticides, exposure, intake conditions, and individuals.

Of real concern are the possible effects of long-term exposure to low levels of DDT and other pesticides. Some sublethal effects have been observed in animals (cellular changes in liver tissue and other physiological and histological effects); however, these effects cannot be extrapolated to man. Of course, there is still the suspicion that DDT might eventually cause damage to human physiology. It has been suggested, for instance, that long-term exposure to low levels of pesticides may cause cancer. Research neither supports nor contradicts this possibility, because it is difficult to control or document experiments on humans over a longer period of time.

Based on animal experiments, the average lethal amount of DDT is a single dose of about 8,000 to 14,000 mg/150-lb person, although quantities as high as 109 ppm have been found in sampling a general population. A study was conducted of persons accidentally or violently killed in Dade County, Florida, from 1965 to 1967. The average concentration of DDT in the fat of the bodies ranged from 5 to 22 ppm; there was more in adults than in children, more in nonwhites than in whites.

Because the use of chlorinated hydrocarbon pesticides has been sharply curtailed, it is therefore expected that smaller quantities of DDT are being ingested and that residues in human fat will decline. However, the average value of DDT, DDE, and isomers in human fat samples in India was 21.8 ± 2.9 mg/kg in 1973 and 24.3 mg/kg in 1965. This would seem to indicate that the DDT storage status had not undergone any significant change in those years.

A comparative study was performed of DDT and its derivatives in human blood samples. The study was conducted in two areas of Ontario where DDT has been used in large amounts. The mean values of total DDT in the adipose tissue and blood samples were found to be 5.83 and 0.032 ppm, respectively. There was a significant correlation between total DDT in the fat and blood, but there was no evidence of any adverse effects of DDT on either of the populations (78).

In another study, tap water samples collected in the Washington, D.C., area before treatment showed the presence of DDT (0.17 mg/ℓ), TDE (0.27 mg/ℓ), and DDT-derived compounds (.15 mg/ℓ); no evidence of these compounds was found after treatment.

The EPA's ban on DDT has been modified since the inception of the pesticide in 1972. In 1974, DDT was used against the tussock moth in northwestern forests and against the pea leaf weevil in Washington and Idaho. The EPA, however, states that DDT will be used only against pests that cannot be controlled by other means and that will do economic damage or threaten human health or safety. Also, despite the suspension of aldrin and dieldrin, the EPA will permit their use against termites and clothes moths, under certain circumstances.

The toxic effects of carbaryl, which is used against DDT-resistant lice, were found to be minimal at the levels of 75 mg/ℓ (135).

The residues of mirex - a chemical which is used to control the imported fire ant - were discovered in the fatty tissue of six persons from 1971 to 1972. It was concluded that the residues came from routes other than direct pesticide usage (478).

The maximum allowable concentration of most of the pesticides (as determined by biological tests) lies below 1 mg/ℓ, but is as low as about 0.01 mg/ℓ for such preparations as atrazine, malathion, and thiometon(522).

Workers occupationally exposed - through manufacturing processes - to aldrin, dieldrin, endrin, and telodrin (isobenzan) for up to 15 yr were studied by Versteeg and Jugar (622). No persistent adverse effects on health were observed in 52 workers who had left the company and were traced. The average pesticide exposure in this group of workers was 6.6 yr, and an average of 7.4 yr had elapsed since exposure. No hepatic disease or convulsions occurred, and no new cases of malignant disease developed. Most of the workers experienced no unusual illnesses.

Exogenous leukemogenic agents such as pesticides (DDT, lindane, organophosphates) are being considered as potentially contributory etiological factors that may activate a latent leukemia virus in certain cells (617).

Organophosphorus Pesticides

The organophosphates attack the neural transmission system of mammals and arthropods and thus interfere with the function of the target organs. Especially active are those organophosphates that function by inhibiting carboxylic ester hydroxylases (including acetylcholinesterase, which is present in human erythrocytes, nerves, and skeletal muscle; and cholinesterase, which is present in human plasma and in the liver). Miosis (contraction of the pupil), is found in about 90 percent of the patients with moderately severe or severe cases of organophosphate poisoning (704).

Tamura et al. (634) conducted a statistical and epidemiologic investigation of the relationship between the changes in the use of organophosphorus insecticides and the recent increase in cases of myopia in 40,000 Japanese children from 1957 to 1973. The study showed that in the year following any period when large amounts of organophosphorus insecticides were used, the morbidity from myopia in school children increased rapidly; conversely, decreased use of the insecticides resulted in decreased morbidity. This it appears that the recent increase in myopia in school children was due, at least in part, to chronic intoxication of organophosphorus insecticides (634).

Pediatric hazards associated with organophosphates are often reported (424). Signs of toxicity are overaction of the parasympathetic nervous system, nausea, vomiting, diarrhea, sweating, and abdominal cramps. Large doses may lead to muscular paralysis and death from respiratory failure (424).

Because organophosphorus pesticides are readily absorbed through the skin, as well as by ingestion and inhalation, these pesticides present a particular hazard to agricultural workers who engage in mixing, loading, and applying the concentrated materials (605).

The incidence of organophosphorus insecticide poisoning varies considerably throughout the world. During 17 yr in Japan there were 19,000 cases of organophosphorus poisoning reported, resulting in more than 9,000 deaths. By contrast, during 17 yr in Great Britain there were only five deaths reported from this poisoning (657). Increased chromosome aberrations were observed in patients suffering acute organic phosphate insecticide intoxication. The frequency of stable chromosome aberrations showed a significant increase with malathion, trichlorfon, mevinphos, and methylparathion; malathion induced an outstandingly high number of structural chromosome aberrations. Patients less seriously intoxicated suffered milder chromosome alterations. Even in the absence of clinical signs of organophosphate poisoning by dichlorvos at low levels, the cholin esterase values were much lower than those of a nonworker control population. The decrease in enzyme levels was significantly correlated with subjective health complaints: sore throat and loss of memory (43).

Herbicides

Today over 40 weed killers are available, but the most widely used are 2, 4-D (2, 4 - dichlorophenoxyacetic acid) and 2, 4, 5-T (2, 4, 5 - trichlorophenoxyacetic acid). In general, these chemicals are rarely lethal to humans or animals, do not persist for long periods of time in the environment, and do not build up in the food chain.

During the Vietnam war, South Vietnam suffered massive military herbicide spraying. It has been estimated that at least one-third of the timber forests was destroyed, as well as 10 percent of all cultivated land and at least 25 percent of the coastal Mangrove forests, which are the breeding or nursery grounds for most offshore fish and crustaceans (686). However, there was no evidence in Vietnamese hospital records that were examined by Thimann (642) that birth defects could be attributed to the herbicide spraying.

Humans are exposed minimally to the phenoxy herbicides through food; air and water are the primary sources of exposure to these herbicides. On the basis of air samples collected from wheat-growing areas in the state of Washington, it was estimated that an average person would be exposed to 1.8 mg of phenoxy herbicide/day. Rain and wind carry the herbicides into water(114). Workmen who were engaged in the manufacture of herbicides were examined; the clinical results were compared to those of a control population that was not exposed to the 2, 4-D or 2, 4, 5-T. No meaningful differences were obtained; moreover, there were no chromosomal effects (114).

In the fall of 1969, the National Institute of Cancer reported that 2, 4, 5-T was teratogenic and was responsible for fetal toxicity at levels of 27 ± 8 ppm. The manufacturer claimed that this herbicide was not teratogenic; that the fetal toxicity was caused by 2, 3, 7, 8-tetrachlorodibenzo-p-dioxin, known as dioxin (333).

At the dose level of 100 mg/kg/day of silvex, there were no effects on either the dams or fetuses of rats. Similar observations were also made with picloram. Picloram, however, is much more persistent in soil than the other compounds. Unlike DDT, it forms soluble salts and does not concentrate in fat; in addition, the toxicity of picloram is very low when compared to that of most other herbicides(333).

The widespread use of phenoxy herbicides has produced no demonstrable evidence of potential harm to man. The herbicides used most widely (2, 4-D and 2, 4, 5-T) are degraded to nontoxic components and do not bioconcentrate (653).

An epidemiological investigation was made of tumor incidence and mortality in Swedish railway workers exposed to various herbicides. Among workers exposed to amitrole (3-amino-1, 2, 4-triazole), tumor incidence and mortality were significantly increased and were slightly dose related. Nearly normal conditions were found in those exposed to phenoxy acids. Animal experiments suggest that amitrole may produce malignant tumors in several different organs, but tumors of the thyroid and liver have received the most attention (23).

To receive a letal dose of 2, 4, 5-T, a 125-lb woman would have to eat the equivalent of her body weight of material containing 380 ppm of the herbicide (based on the acute oral toxicity expressed in mg/kg). Both 2, 4, 5-T and DDT in technical form are relatively nontoxic on skin contact (3,800 mg/kg for 2, 4, 5-T and 2,500 mg/kg for DDT). There have been no human deaths and remarkably few human illnesses from the agricultural or public health uses of either of these chemicals. A no-effect level of 50 mg/kg/day of 2, 4, 5-T containing less than 1 ppm dioxins is proposed as providing ample protection for human embryos (114).

The first recognized incident in which significant poisoning resulted from the improper disposal of waste residues containing TCDD (tetrachlorodibenzodioxin) was in eastern Missouri. In this incident, a salvage oil company sprayed waste-oil sludge on an arena at a horse-breeding farm, to control dust. Birds, cats, dogs, and rodents were killed; 62 out of 85 horses became ill, and 48 died. The horses had been exposed to the arena in the summer of 1971, and they continued to die as late as January 1974. The horses showed chronic weight loss, loss of hair, skin lesions, dependent edema, intestinal colic, dark urine, gross hematuria (the passage of blood in the urine), conjunctivitis, joint stiffness, and laminitis (inflammation of one side of the neural arch of a vertebra). In addition, the horses' feet were inflamed. Human illness, although less severe, included one case of hemorrahagic cystitis in a 6-yr-old girl who played in the arena. The soil of the arena contained 3.18 to 3.3 percent TCDD by weight and 2, 4, 5-tri chlorophenol (TCP) and related chemicals (662).

Paraquat, a widely used bipyridyl herbicide, produces a low-dose, chronic illness in rats, primarily manifested by pulmonary fibrosis (216). Paraquat is also known to cause severe lung damage in rats, leading to death. Although it causes proliferation of fibroblasts, no carcinogenic action has been demonstrated. Taken orally, paraquat causes ulceration in the digestive tract, diarrhea and vomiting, renal damage, and jaundice.

Pesticidal Viruses

Tinsley and Melnick (645) found some antibodies that reacted with insect viruses in domestic and wild animals and in several laboratory workers handling insect viruses. There is always a possibility that changes in the pathogenicity and specificity of pesticidal insect viruses could occur, causing a wider spectrum of host involvement. No collaborative research programs on the in vitro specificity of insect viruses were recommended (645).

Fungicides

In Iraq during a two-month period, 6,530 poisoning victims were hospitalized, and 459 hospital deaths occurred. The source of the poisoning was found to be homemade bread prepared from seed wheat treated with methyl mercurial fungicides (634). In another case, diphenyl (or biphenyl) poisoning from fungicide in a Finnish paper mill was reported by Seppäläinen and Häkkinen (573). Workers were employed in areas of the mill where the average concentration of diphenyl measured in the air varied from 0.6 to 123.0 mg/m^3. The workers developed EEG abnormalities that were compatible with generalized cerebral disturbance (573).

Polychlorinated Biphenyls (PCB's)

Relatively high concentrations of a group of widely used industrial chemicals known as PCB's have been found in fish, birds, and man. The widespread presence of these PCB's has tagged them, like DDT, as truly global pollutants. In fact, it has been speculated that sunlight might convert DDT to PCB's. PCB's are insoluble in water, soluble in fats and oils, and very resistant to chemical and biological degradation. Due to their solubility, PCB's accumulate in the environment, especially in aquatic organisms and birds (in which cumulation factors up to one billion may be reached). PCB's, like DDT, can inhibit photosynthesis of marine phytoplankton and can kill shrimp, trout, minks, and birds. PCB's may be twice as effective as DDT in causing thinning of bird eggshells.

In 1968, over 1,000 people in Japan suffered from a skin disease and from liver damage caused by rice oils. The oil was heavily contaminated with PCB's; however, these effects were not due to the PCB's but to a highly poisonous contaminant - chlorinated dibenzofurans. This contaminant is found in some PCB's manufactured in other countries.

To date limited study in this area indicates that uncontaminated PCB's have a very low toxicity to man. According to the Food and Drug Administration, the average PCB concentration in a normal American diet is only about 10 percent of the strict safety levels set in 1971 for food, food packaging materials, and animal feeds.

SYNTHETIC/ORGANIC CONTAMINANTS

Recently diverse compounds identified in water supplies drawn from the Mississippi River have been discovered in the blood serum of local residents using the water supply. This has created great concern over chemicals found in drinking water. The presence of small amounts of synthetic/organic chemicals in treated reclaimed water has been recognized as a potential health hazard.

The list of compounds identified in drinking waters is rapidly growing larger. This is due primarily to the continual introduction of new chemicals but also to the development of sensitive analytical techniques that measure trace quantities of the chemicals. There is very little evidence available concerning the relation between the presence of these compounds in water and human disease. Information on classical acute health effects of relatively toxic chemicals can be obtained from physicians' manuals. However, knowledge of the chronic health effects associated with long-term exposure to low-level concentrations of chemical substances is not well documented. The possibility that cancer may result from long-term exposure to low concentrations of carcinogens is of utmost concern.

Carbon-Chloroform Extractables (CCE) and Carbon Alcohol Extractables (CAE)

The Committee on Water Quality Criteria (672) suggested that absorbable organic carbon in public water supply sources should not exceed the carbon chloroform extractables (CCE) level of 0.7 mg/ℓ. (No level has been established for carbon alcohol extractables - CAE). The establishment of this level was based upon the adverse physiological effects of CCE as well as aesthetic considerations.

To date, laboratory testing of the epidemiological and pathological effects of trace organics has been restricted to mice and fish. Hueper and Payne (306) conducted a study of mice that were exposed subcutaneously, cutaneously, and orally to extracts of CCE and CAE obtained from both raw and finished water supplies. Results of this study indicated that these extracts had a potential for carcinogenicity (see Table 99). The cutaneous dose used in the experiment was one drop of extract every 2 weeks for 56 weeks with 72 mice; the subcutaneous dose was 2 to 4 mg every 2 weeks for 56 weeks with 72 mice; and the oral dose administered as 2 percent of the raw powdered food was eaten ad libitum by the animals for a 13-month period during the study. The control group consisted of 40 unexposed mice. No tumors were observed among the 40 mice in the control group or among those exposed orally. Carcinogenic effects in subcutaneous and cutaneous groups included one papilloma (a circumscribed overgrowth of the papilla) of the bladder; four spindle-cell sarcoma (fusiform cell tumor) at the site of subcutaneous injection; and leukemia, lymphoma, or reticulum-cell sarcoma of the liver in all other instances. However, it is very difficult to extrapolate the experimental data of animals to humans. Thus, the question still remains as to whether such extracts would cause similar abnormalities in humans.

Another experiment was conducted to investigate the toxicity of CCE and CAE from processed water supplies (187). A total of 5 mg of either CCE or CAE was introduced subcutaneously during the first 20 days after birth of the test mice. No tumors were found to be induced during the period, but varied death rates of the test animals were observed. The studies were conducted over a 1-yr period using New Orleans drinking water supplies (187).

Organohalides

Occurrence and formation of organohalides such as $CHCl_3$, $CHCl_2Br$, $CHClBr_2$, and $CHBr_3$ were reported when water containing organic substances was chlorinated (41, 554). Of the haloforms, chloroform ($CHCl_3$) was reported as the predominant organohalide, with concentrations ranging from 54 μg/ℓ to 150 μg/ℓ. The level of risk for chloroform - estimated from consideration of the worst case and for the expected cancer site, such as the liver - might be extrapolated to account for up to 40 percent of the observed liver cancer. The toxicity of chloroform has been well demonstrated in lethal-dose studies.

TABLE 99 . SUMMARY OF RESULTS OF INTRODUCING CCE AND CAE FROM RAW AND FINISHED WATER INTO MICE (306)

Route of Exposure	Number of Tumors Produced/Animal Exposed			
	Water Source			
	Raw		Finished	
	CCE	CAE	CCE	CAE
Subcutaneous	5/72	1/72	2/72	1/72
Cutaneous	2/72	1/72	0/72	0/72
Oral	--*	--	0/40	--

* No test.

An LD_{50} (lethal dose - 50 percent) value ranging from 89 to 35 mg/kg was observed by Tardiff and Deinzer (636) when CCE obtained from the Kanawha River in West Virginia was introduced into mice via an interperitoneal route. The differences among these LD_{50} values was shown to be due to the amount of chloroform present in the extract, indicating the toxicity of chloroform. The CAE obtained from the same river showed an LD_{50} of 84 mg/kg. The LD_{50} for the concentrated organics from Cincinnati tap water was shown to be 65 to 290 mg/kg.

The same authors also reported the identification of 60 compounds from drinking water. Of the compounds, 1 was classified as nontoxic; 14, moderately toxic; 16, very toxic; 2, extremely toxic; and 27, unknown (74). However, it is difficult at this time to determine the relationships of the toxicities of these compounds in humans to the level of the compounds present in water and wastewater.

Careful interpretation of the toxicity data, such as the LD_{50} value obtained from concentrated extracts, is necessary when these values are to be used to set toxicity levels in drinking water.

Polynuclear Aromatic Hydrocarbons (PAH)

Occurrence, formation, concentration, activity, carcinogenicity, and degradation of polynuclear aromatic hydrocarbons (PAH) in water are well documented (17).

Of the PAH, 3, 4-benzpyrene has been generally recognized as the most potent carcinogen. Minimal carcinogenic doses of three of the most potent hydrocarbons in susceptible experimental animals is shown in Table100 .

TABLE100 . THE MINIMAL CARCINOGENIC DOSE FOR THREE OF THE MOST POTENT CARCINOGENIC HYDROCARBONS IN SUSCEPTIBLE EXPERIMENTAL ANIMALS (716)

Carcinogen	Animal	Least Amount which Caused Cancer*
3,4 - Benzpyrene	Mouse	4.0 μg
3,4 - Benzpyrene	Rat	50.0 μg
1,2,5,6 - Dibenzanthracene	Mouse	2.5 μg
20 - Methylcholanthrene	Mouse	4.5 μg
	Rat	20.0 μg

* Only one dose was administered subcutaneously.

Miscellaneous Organic Compounds

A preliminary experiment was designed to study the toxicity of organic compounds present in a secondary treatment plant effluent. Rats were supplied with filter-sterilized effluent from an activated sludge plant as the sole source of drinking water. Two of the 10 rats developed massive tumors. Also, the exposed female rats developed significantly smaller adrenal glands than the control rats that were provided with the local water supply (496).

An epidemiologic study on the toxicity of compounds present in drinking water was cited by Andelman and Suess (17). This study indicated fewer cancer mortalities in a London borough that was supplied with well water than in boroughs supplied with river water. This could mean that the river water receives more carcinogenic waste material.

Similar findings reported by Tromp (650) showed that areas using municipal water systems had lower cancer death rates than those using other systems. However, the cancer death rate was higher among areas that received municipal water from a river than among those that received municipal water from wells.

When other factors such as food, air quality, and individual habits (i.e., cigarette smoking) are considered, the importance of trace carcinogens in water supplies may not be significant. However, several observations have been made correlating water supply quality and cancer incidence. For example, Talbot and Harris (633) established a correlation between cancer mortality in white males and water supply source, between mortality and urbanization, and between mortality and income. When occupational variables are not considered, lung cancer mortality rates were found to be correlated with surface water sources, but there were no correlations found in other cancers.

Halogenated Hydrocarbons

In a study that was designed to investigate the correlation between levels of halogenated hydrocarbons in New Orleans drinking water and levels of halogenated hydrocarbons in blood plasma of individuals drinking the water, 13 halogenated hydrocarbons were isolated. Researchers also detected the presence of carbon tetrachloride and tetrachloroethylene (2 of the 13 compounds) in the pooled sera of eight people. It is probable that biomagnification was involved if the chemicals in the plasma originated from the drinking water (180).

Low Molecular Sulfurated Hydrocarbons

A case history study of the waterborne goitrogens and their role in the etiology of endemic goiter was recently reported from Colombia, South America (229). The potential presence of low molecular weight compounds (less than 220), such as sulfurated hydrocarbons in water and wastewater, received careful evaluation. The compounds (sulfurated hydrocarbons) were known to be related to the high incidence of goiter among children and were regarded as waterborne goitrogens. The study also reported a 10-fold increase in cancer of the thyroid where endemic goiter was observed (229).

BIOLOGICAL CONTAMINANTS

Epidemics of waterborne diseases have largely been eliminated, due mainly to the advancement of sanitary engineering, enforcement of public health regulations, and preventive medical practices; however, waterborne disease data from the last three decades indicate that outbreaks are no longer on the decline in the United States. During the 25-yr period from 1946 to 1970, there were 358 recognized outbreaks (72,358 individuals involved) of disease or chemical poisoning attributed to contaminated drinking water (145, 220).

According to the reports of the Center for Disease Control (221) during the past four years (from 1972 to 1975), 105 waterborne disease outbreaks were reported, involving 22,650 cases. As shown in Table 101, in 1975, 24 waterborne disease outbreaks were reported, involving 10,879 cases.

TABLE 101. WATERBORNE DISEASE OUTBREAKS
1972-1975 F AND W (145)

	1972	1973	1974	1975	Total
Outbreaks	29	24	28	24	105
Cases	1,638	1,720	8,413	10,879	22,650

Table 102 shows the number of outbreaks and cases by etiology and type of water system. The category with the most outbreaks is acute gastrointestinal illness. This category includes outbreaks characterized by upper and/or lower gastrointestinal symptomatology for which no specific etiologic agent was identified. In previous years, these outbreaks were considered sewage poisoning. One outbreak each was caused by giardiasis, shigellosis, enterotoxigenic E. coli, and hepatitis A. There were no reported deaths associated with waterborne disease outbreaks in 1975 (221).

Most outbreaks involved semipublic (67 percent) and municipal (25 percent) water systems; some involved individual (8 percent) systems. Outbreaks attributed to water from municipal systems affected an average of 1,218 persons; those attributed to semipublic systems involved 221 persons; and those associated with individual water systems affected 13 persons. Of those 16 outbreaks associated with semipublic water supplies, 11 (69 percent) involved visitors to areas used mostly for recreational purposes.

TABLE 102. WATERBORNE DISEASE OUTBREAKS, BY ETIOLOGY AND TYPE OF WATER SYSTEM, 1975 (221)

	Municipal		Semipublic		Individual		Total	
	Outbreaks	Cases	Outbreaks	Cases	Outbreaks	Cases	Outbreaks	Cases
Acute gastrointestinal illness	4	7,300	13	2,460	--	--	17	9,760
Chemical poisoning	2	11	1	26	--	--	3	37
Giardiasis	--	--	--	--	1	9	1	9
Shigellosis	--	--	1	56	--	--	1	56
Enterotoxigenic _E. coli_	--	--	1	1,000	--	--	1	1,000
Hepatitis	--	--	--	--	1	17	1	17
Total	6	7,311	16	3,542	2	26	24	10,879

The object of this section is to compile a comprehensive summary of research into the health effects of biological contaminants in wastewater treatment. Diseases transmitted by water and wastewater largely originate in the intestinal discharge of man and/or animals. These diseases are caused by bacteria, viruses, fungi, and protozoan and other parasites.

Protozoan and Other Parasites

A number of intestinal parasite infections can be introduced into man directly from water supplies and indirectly through wastewater discharges. Under normal conditions, the potable-water route of infection is quite unimportant. However, the reuse of treated waste effluents for potable purposes requires that this problem be reexamined.

Ascariasis (a disease caused by infection with ascaris), trichuriasis (a disease caused by infection with trichuris), and hookworm diseases are some of the infections that originate from direct soil pollution by feces. This pathway has been virtually eliminated in the United States, due to the introduction of modern sewage disposal and water supply systems. When there is a breakdown in sanitation, these diseases may reappear (699).

Studies in foreign areas affirm the relative unimportance of public water supplies as a route of infection for intestinal parasites. A study by the World Health Organization (WHO) in Sudan, in which a modern water supply was provided to one test city, showed that parasitic infections were not decreased. The study indicated, instead, the need for sanitary waste disposal facilities. This finding was substantiated by the substitution of a modern water supply system for an older system in Western Transvaal. The substitution had no effect on the prevalence of helminths among the Bantu population studies - children 7 to 16 years (699).

Amoebic dysentery (or amebiasis) appears to be the most important parasitic disease associated with wastewater in the United States. It is caused by Entamoeba histolytica, a protozoan. Today, the prevalence rate of E. histolytica in the general population of the United States is considered to be around 3 to 5 percent (383). The prevalence of the intestinal protozoa varies considerably in different population groups and is generally correlated with socioeconomic conditions. Higher rates are found in areas of poor sanitation and in regions without sewage systems and potable water. Higher rates are also noted in groups of people with poor personal hygiene (e.g., patients in institutions for the mentally retarded).

The amoeba can form small cysts (5 to 20 μm) with a specific gravity of about 1.06. Each mature cyst is capable of producing four motile amoebae. The cysts are resistant to adverse environmental conditions and are excreted with feces into water and/or remain in the human digestive tract to become vegetative amoebae. These amoebae multiply and may become invasive, causing erosion of the superficial mucous membranes. They may eventually invade the tissue with consequent ulceration.

The vegetative forms do not survive outside the digestive tract. As with most parasitic diseases, the symptomatology produced by pathogenic intestinal protozoa is too nonspecific to enable the physician to make an accurate clinical diagnosis. In 1974, there were 2,743 reported cases of amebiasis in the United States (458), because of nonclinical manifestations, the actual figure is undoubtedly considerably higher.

In an experiment with volunteers, it has been demonstrated that up to 25 percent could be infected by a dose containing less than 10 organisms of entamoeba; the remainder required a minimal dose of 10,000 organisms to become infected. However, as shown in Table 103, the infected volunteers did not manifest any signs of illness.

Giardia lamblia, a flagellated protozoan of the small intestine, often implicated epidemiologically with drinking water is the etiological agent for giardiasis. An outbreak has recently been reported (460) in Rome, New York, where the water supply could have been contaminated by untreated human waste. Another outbreak of giardiasis by G. lamblia was reported in September 1976 in Idaho. The source was purported to be from untreated surface water of an individual water system (290).

Apparently, the cysts of G. lamblia survive in water and remain infective for 16 days. During 1969 to 1973, seven outbreaks involving 193 people were reported in the United States. During October 1954 to March 1955, there was a suspected waterborne outbreak of 50,000 cases of giardiasis in Portland, Oregon. The outbreak was not reported, because of the failure to isolate the organism from the suspected water source (451). Epidemic giardiasis among American travelers to the Soviet Union has been reported since 1970; the latest outbreak was reported in October 1975 (460). Sporadic single cases or occurrences of giardiasis with recent exposure to untreated mountain or pond water have been noted (457). In an experiment where adult humans were given challenge doses of Giardia lamblia, 76 to 100 percent infected with a dose containing 10 organisms did not become ill. Similar results were observed with doses containing up to a million organisms (see Table 103).

An outbreak of ascariasis occurred after World War I; one out of every three surgical patients at a hospital in Le Havre, in the 2 yr following cessation of the war, was found to have the disease (699). Also, following World War II, a 40 percent incidence of ascariasis was reported in Darmstad and was attributed to a widespread breakdown of sanitation practices that occurred in Germany during the latter part of the war (699).

In moderate climates, the human contribution of ova to wastewater would appear to be no greater than 10 percent, but may reach 30 percent in subtropical regions such as the southern extremities of the United States. The remainder of the ova is of animal origin. Various authors have reported 59 to 80 worm eggs/ℓ sewage (223). The eggs are generally resistant to environmental conditions, having a thick outer covering to protect them against desiccation. In one study, 90 percent of ascaris ova was destroyed after 15 days at 29°C; the ova may survive for up to 60 days at 40°C (223).

Ova from the giant roundworm Ascaris lumbricoides, the pinworm Oxyuris vermicularis, the whipworm Trichuris trichuria, the tapeworm Taenia saginata, and possibly the hookworm are

TABLE 103. CLINICAL RESPONSE OF ADULT HUMANS TO VARYING CHALLENGE DOSES OF ENTERIC PATHOGENS (88)

ORGANISM	Challenge doses											
(strain)	10^0	10^1	10^2	10^3	10^4	10^5	10^6	10^7	10^8	10^9	10^{10}	10^{11}
Shigella dysenteriae												
(1) (A-1)			+		++							
(1) (M 131)		+	++	+++	++++							
Shigella flexneri (w)										+++		
(2a)					+	+++	++++	+++	++++			
(2a)			+	+++	+++	+++						
Vibrio cholerae												
inaba 569B (unbuffered)					-		-	-	++	++	-	++1
inaba 569B (+NaHCO$_3$)		-		(+)	+++	+++1	+++1		++1			
ogawa (+NaHCO$_3$)				++			++1					
Salmonella typhi												
(Quailes)vi				-		++		++	++++	++++		
(Zermat)vi					+++							
(Ty2V)vi							++					
(0-901)									++			
(Ty2W)									+			
(Quailes)						+		++		++++		
(Quailes)						++						

TABLE 103. (continued)

ORGANISM	Challenge doses											
(strain)	10^0	10^1	10^2	10^3	10^4	10^5	10^6	10^7	10^8	10^9	10^{10}	10^{11}
Salmonella newport						+	++					
Salmonella bareilly						+	++					
Salmonella anatum (I)					-	+						
(II)					-	-	-	+				
(III)					-	-	++	+	+			
*							++	+	+			
Salmonella meleagridis (I)					-	-	-	++				
(II)							-	++				
(III)						-	+	++				
*								+++	+++			
Salmonella derby						-	-	++				
Salmonella pullorum (I)					-	-	-	-	-	-	++++	
(II)					-	-	-	-	-	++++		
(III)					-	-	-	-	-	++++		
(IV)					-	-	-	-	-	++		

TABLE 103. (continued)

ORGANISM	Challenge doses											
(strain)	10^0	10^1	10^2	10^3	10^4	10^5	10^6	10^7	10^8	10^9	10^{10}	10^{11}
Escherichia coli (0111:B4)							+++		+++	+++		
(055:B4)									+++	+++	++++	
(06:H16)									++		+++	
(0124:K72:H-)									+		(+)	
(0143:K?:H-)					(+)		(+)		+++			
(0144:K?:H-)					-		+		+++			
(0148:H28)									+		++++	
Streptococcus faecalis var. liquefaciens									-	+	++	
Clostridium perfringens type A (Heat-resistant)									++	+++		
Clostridium perfringens type A (Heat sensitive)										++++		
Endamoeba coli	(+)	(++)	(++)	-	(++++)							
Giardia lamblia	-	(++++)	(++)		(++++)	(++++)	(++++)					

TABLE 103. (continued)

ORGANISM	Challenge doses											
(strain)	10^0	10^1	10^2	10^3	10^4	10^5	10^6	10^7	10^8	10^9	10^{10}	10^{11}
Norwalk agent (virus)1 ++ ++ 2 +++ 3 +												
Hepatitis virus A +++ (fecal ++ viltrates)												

- = 0, + = 1-25, ++ = 26-50, +++ = 51-75, ++++ = 76-100 percent of volunteers developing illness.

* Refeeding trials of volunteers who mo before became infected by the same strain.

** 1,2,3 refers to serial passages of stool filtrates.

(+) Infections without illness.

1 = cholera-like diarrhea.

reported to be present in wastewater (223). Heavy infestations of roundworms found in some European cities are related to the use of night soil, which is also known to be responsible for about 20 percent of the recurrent infections of amoebiasis and hookworms (237).

Man is known to be the host and reservoir of A. lumbricoides, whose ova are excreted in the feces of infected individuals. Ova of these intestinal parasites require several days before maturing to an infective stage. Under the most favorable conditions, ascarids require 10 days to mature; trichurids, 21 days; and hookworms, 6 days. Hence, these parasites are unlikely to pose health threats if the water is used for drinking, since they would be eliminated from the human body before maturation. However, use of such water for food processing, garden watering, or simple aesthetic activities may require the effective removal or destruction of the parasites (699).

Free-living nematodes are widely found in municipal water supplies. Their potential as carriers of enterococci, salmonella, and shigella has been demonstrated (699). Although free-living nematodes are not important as health threats in conventional waterworks practice, their significance in a reclaimed water situation needs evaluation.

Dracontiasis, or guinea worm infection, is a disease primarily associated with poverty (especially inadequate water supplies and wastewater treatment), and is common in India and West Africa. Vector species of Cyclops persisting in ponds used for drinking water must be controlled (usually by the use of Abate at a concentration of 1 mg/ℓ).

In summary, it may be stated that a large quantity of a variety of ova from parasitic worms may be present in wastewater, and that the ova possess a high degree of resistance to many environmental stresses.

Fungi

Candida albicans, a pathogenic, yeastlike fungus, is an asporogenous (non-spore-forming) yeast that develops pseudohyphae (a kind of filamentous structure). Large, round, thick-walled chlamydospores - the morphological characteristic of the genus candida - are frequently, though not always, present in wastewater.

C. albicans has been found in the feces and skin of several animal species other than man. The fungus rarely occurs in soil. The prevalence of candida in feces in different regions of the United States varies from 19.3 to 44.6 percent of the total population (323). Seventy percent of the inhabitants of Baghdad have the fungus in their feces. Another important infective area in the human body is the female vagina; 43 percent of the female population in the United States are carriers (323).

C. albicans may cause oral thrush, corneal ulcers, and other ocular infections. One survey analyzed 20 weekly samples collected at three stations on the North Shore of Great South Bay, Long Island. It was reported that the estuarine water samples contained between 1,000 to 11,000 or more cells/ℓ (323). These results are shown in Table 104.

TABLE 104. AVERAGE VALUES FOR C. ALBICANS, COLIFORM AND FECAL COLIFORM COUNTS AND TOC (TOTAL ORGANIC CARBON) DETERMINATIONS IN THE ESTUARINE WATER SAMPLES, LONG ISLAND, NEW YORK (323)

Estuarine Stations	*C. albicans* cells/ℓ	Coliform Total MPN/100 mℓ	Coliform Fecal MPN/100 mℓ	TOC mg/ℓ
Station 1	4,245	338	33	8
Station 2	4,003	298	12	8.4
Station 3	9,555	1,962	490.5	8.7

This fungus retained an infectivity and pathogenicity to mice after it was exposed to sea water for eight weeks (323). In the open ocean, there is a concentration of 200 to 300 fungal cells/ℓ; in moderately contaminated beach areas, there are levels of 10,000 to 20,000 cells/ℓ; and in heavily contaminated estuaries, up to 100,000 cells/ℓ (323). The overall trend was a gradual increase in concentrations during summer months, from June to September, after which the concentration declined (323).

Bacteria

Salmonellosis--

A wide variety of species that are pathogenic to man and animals belongs to the genus Salmonella. Water and food, as well as personal contact, are the main routes for transmission of the species from man to man.

Among the three distinct forms of salmonellosis in man, typhoid fever - caused by *S. typhi* - is the most severe enteric fever form, and man is the only host. Salmonella septicemia, most commonly caused by *S. choleraesuis*, is relatively rare. This bacteria is not particularly common in humans, but has a predilection for swine. Salmonella are most commonly encountered in acute gastroenteritis. Serotypes (types based on antibodies) in excess of 1,500 have been identified. Most, in contrast to *S. typhi*, are not host specific.

The death rate, due to typhoid fever in the United States in 1900, was 31.3/100,000 population; however, at present, death due to this disease is practically nonexistent, as shown in Table 105 (164, 308).

TABLE 105. U.S. MORTALITY FROM SELECTED CAUSES RELATED TO WATER POLLUTION (164)

	Death Rate per 100,000					
Cause of Death	1920	1930	1940	1950	1960	1967
Typhoid & paratyphoid fever	7.6	4.8	1.1	0.1	0.0	0.0
Dysentery	4.0	2.8	1.9	0.6	0.2	0.1
Gastritis, duodenitis, enteritis, & colitis	53.7	26.0	10.3	5.1	4.4	3.8

Relatively few outbreaks of typhoid fever and salmonellosis associated with drinking water were reported in the United States during 1971 to 1973 (220, 448). The reported isolation rates for humans in the United States in 1972 was 12.5/100,000; the fatality rate between 1962 and 1972 was 0.43 percent, mostly among the very young and very old.

In 1974, 23,833 isolations of salmonella were reported to the Center for Disease Control, a decrease of 2,855 cases (10.7 percent) from the previous year. As in 1973, S. typhimurium (30.8 percent), S. newport (6.9 percent), and S. enteritides (6.0 percent) were the first, second, and third most commonly isolated serotypes, respectively (563). The annual incidence of reported human isolations of salmonella has remained relatively constant since 1963 (563).
The seasonal incidence for the period from 1967 to 1974 shows a consistent pattern, with the greatest number of isolations reported in July through November, and the fewest reported in February through April (563).

The ages of infected persons reveal that 66.8 percent of the 17,229 isolations was from persons less than 20 yr of age. Similarly, this age group showed the highest infection incidence for the years from 1963 through 1973 (563).

Several serotypes (e.g., S. weltevreden, S. panama, and S. oslo in Hawaii; S. newport and S. javiana in the southern states) have definite regional patterns, for reasons that are not clear.

From 1962 to 1974, 143 deaths were reported among the 34,291 persons involved in 499 outbreaks. This resulted in a case-fatality ratio of 0.42 percent.

In 1974, in 18 of the 34 outbreaks of salmonellosis (involving a total of 4,011 persons), 5 outbreaks were caused by contaminated poultry, 5 by beef or beef products, and 3 by dairy products. In 6 of these outbreaks, person-to-person transmission was thought to be responsible; none was reported to be transmitted by water. In 1973, 3 typhoid outbreaks (totalling 217 cases) and 1 outbreak of salmonellosis (3 cases) occured in semipublic or individual water supplies. In 1974, an outbreak involving several hundred people on a cruise was reported; epidemiologic investigation failed to clearly implicate either food or water. Three epidemiologic investigations of turtle-associated salmonellosis were also found in the literature(144).

According to experimental data, the dose required to bring about human cases of typhoid fever is surprisingly high (300). Human volunteers were challenged with various doses of _Salmonella typhi_. The results are summarized in Table 106.

TABLE 106. RELATION OF DOSAGE OF _S. TYPHOSA_ TO DISEASE (300)

Number of Viable Cells _S. typhosa_	Total Volunteers Challenged	Number with Disease
10^9	42	40 (95%)
10^8	9	8 (89%)
10^7	32	16 (50%)
10^5	116	32 (28%)
10^3	14	0 (---)

With the salmonella species isolated from spray-dried eggs, human volunteers were challenged orally; the results are summarized in Table 107. Existence of varying degrees of virulence among the species and strains are shown. Similar results are shown in Table 103.

TABLE 107 . DOSE OF VARIOUS SPECIES AND STRAINS OF SALMONELLA THAT CAUSED DISEASE IN HUMAN VOLUNTEERS (142

Salmonella Species/Strain	Dose at which 50% or More Develop Clinical Disease
S. meleagridis I	50,000,000
S. meleagridis II	41,000,000
S. meleagridis III	>10,000,000
S. anatum I	860,000
S. anatum II	67,000,000
S. anatum III	4,700,000
S. newport	1,350,000
S. derby	15,000,000
S. bareilly	1,700,000
S. pullorum I	>1,795,000,000
S. pullorum II	>163,000,000
S. pullorum III	>1,295,000,000
S. pullorum IV	1,280,000,000

Shigellosis--

Shigella cause bacillary dysentery in man and in higher apes. Although person-to-person transmission is the predominant mode of spreading shigellosis, waterborne outbreaks have played a significant role in the overall epidemiology of the disease in the United States(579, 612). In 1975, 14,757 shigella isolations from humans were reported to the Center for Disease Control (CDC). This was a decrease of 24.0 percent from the 19,420 isolations reported in 1974 (580). Utilizing population estimates for July 1, 1975, approximately 69.2 isolations were reported for each million population of the United States in 1975. *Shigella sonnei* (60.3 percent) was the most common etiological agent in all these cases, followed by *S. flexneri* (38.2 percent). Between January 17 and March 15, 1974, approximately 1,200 cases of acute gastrointestinal illness occurred in Richmond Heights, Florida. The outbreak was caused by a failure in the chlorination process of well water, which allowed insufficiently chlorinated water from a contaminated well (located near a church's septic tank) to be distributed to the community (144).

Most instances of Shigella-induced illnesses reported in the past several years have involved small wells, temporary breakdowns of chlorination systems in water supply, and swimming in waters contaminated with sewage. The isolation rate in the United States is approximately 15/1,000,000 population. Up to 25 percent of adult humans may be infected and show clinical response to Shigella dysenteriae doses of 10 cells; 25 to 50 percent, to doses of 100 cells; and up to 100 percent, to doses of 10^4 cells (see Table 103). These results indicate that it would take smaller doses to show clinical response for shigella than it would take to show response for salmonella.

Cholera--

Cholera, which is fully controlled in the United States, has created a major global public health problem in India, Italy, Portugal, and many other countries. In the United States, only one case (in the Gulf Coast town of Port Lavaca, Texas) has been reported since 1911. Transmission was associated with a well that was contaminated by leachate from a septic tank system.

The etiological agent of cholera is Vibrio cholerae, Biotype El Tor, Serotype Inaba or Ogawa. Man is the only known natural host, and, since a prolonged carrier state is uncommon, the disease must be maintained by an unbroken chain of mild, subtle infections. Cholera is a serious disease, similar to typhoid fever but more rapid in onset, more virulent, and more often fatal. Death rates of 25 to 85 percent are commonly reported (142).

The primary means of cholera transmission is the drinking of contaminated water. Also associated with cholera is the eating of fish caught in contaminated waters. In the event that imported cases occurred in the United States, it is felt that the risk of spread would be minimal because of modern, industrialized sanitary engineering as well as responsible medical therapy. It is also reported that vaccination is not needed to control imported cases or outbreaks that may occur in the United States (144).

The minimum doses of vibrio that cause clinical symptoms in adult humans may vary (depending upon the strains of vibrio that are used) from 10^3 cells (in up to 50 percent of the individuals) to 10^5 cells (in up to 75 percent of the individuals), as shown in Table 103.

Gastroenteritis--

There are many reports of waterborne gastroenteritis of unknown etiology in which bacterial infections are suspected. These include outbreaks characterized by nausea, vomiting, diarrhea, and fever, for which no specific etiologic agent could be identified.

An epidemiological study of the impact of wastewater pollution on marine bathing beaches was conducted during the summers of 1973 and 1974 at the Coney Island and Rockaways beaches in New York (95). A statistically significant finding was observed: the rate of gastrointestinal symptoms among swimmers compared to nonswimmers was higher at Coney Island, where the densities of the indicative organisms such as E. coli and fecal streptococci were significantly higher than the densities at Rockaways Beach (Tables 108 and 109). It was concluded that there were measurable health effects associated with sewage polluted waters (95).

In 1971, a waterborne gastroenteritis outbreak was reported in Pico Rivera, California, in which 11,000 residents became ill with diarrhea and abdominal cramps. No pathogens were isolated from any cases. The source of water was responsible for the outbreak; chlorination at the reservoir had been interrupted when the chlorine supply was exhausted. One of the major outbreaks involving over 1,000 persons occurred at Crater Lake National Park, Oregon, in July 1975. The illness was reported to be associated with sewage-contaminated water (459). Enterotoxigenic E. coli, Serotype 06:H16, was isolated from ill park residents and from the park's water supply (221).

It was noted that the attack rate of acute "summer diarrhea" on the Fort Apache Reservation, Arizona, during 1971, rose simultaneously with rainfall, temperature, and bacterial contamination of water sources (703). Nonenteropathogenic E. coli capable of producing enterotoxin was isolated.

In the study of clinical response of adult humans to challenge doses of E. coli, it has been demonstrated that very large doses of cells (about 10^8) would be necessary to show clinical response in 75 percent of the individuals tested (Table 103). With the E. coli strain of 0111:84, doses of only 10^6 cells were necessary for a similar response. However, it should be noted that the digestive tract in most individuals is populated by normal flora, of which E. coli is the most abundant and most characteristic; about 10^9/g of feces is common(44).

TABLE 108. MEAN INDICATOR DENSITIES AT THE CONEY ISLAND AND ROCKAWAYS BEACHES, NEW YORK, DURING 1973 AND 1974 TRIALS (95)

Indicator	Log Mean Recovery/100 mℓ			
	1973		1974	
	Coney Isl.	Rockaways	Coney Isl.	Rockaways
Total coliforms	983*	39.8	1213*	43.2
Fecal coliforms	165*	21.5	565*	28.4
Escherichia coli	174*	24.8	15.3*	2.4
Klebsiella	112*	13.7	59.2*	3.5
Enterobacter-citrobacter	530*	11.1	434	6.6
Fecal streptococci	91.2	21.8	16.4*	3.5
Pseudomonas aeruginosa	30.4	6.5	45.8*	3.1
Aeromonas hydrophila	25.3	26.5	9.6	4.9
Vibrio parahaemolyticus	ND	ND	54.5	32.8

*Significantly different at 95 percent confidence level.

TABLE 109 . SYMPTOM RATES IN PERCENT AT CONEY ISLAND AND ROCKAWAYS BEACHES, NEW YORK, DURING 1973 AND 1974 TRIALS(95)

Symptom Group	Rates in Percent for Symptom Groups in											
	1973						1974					
	Coney Island			Rockaways			Coney Island			Rockaways		
	S	NS	×	S	NS	×	S	NS	×	S	NS	×
N	474	167		484	197		1961	1185		2767	2156	
Resp.	12.9	10.2	2.7	18.0[ab]	11.7	6.3	7.2	6.4	0.8	8.3	7.8	0.5
G.I.	7.2[a]	2.4	4.8	8.1	4.6	3.5	4.2[a]	2.6	1.6	3.9	3.5	0.4
Other	9.9	6.6	3.3	9.1	8.6	0.5	7.3	6.7	0.6	8.6	7.7	0.9
"Severe"	5.9	4.2	1.7	6.0	5.6	0.4	3.8	2.9	0.9	3.0	2.6	0.4

a Significantly (P = 0.5) higher than nonswimmers.

b Significantly (P = 0.5) higher than other beach.

N-sample size; S-swimmers; NS-nonswimmers; ×-difference

GI - gastrointestinal; Resp. - respiratory

Leptospirosis--

Etiological agents of leptospirosis (an infection caused by leptospira) constitute approximately 150 different serotypes categorized on the basis of their agglutinogenic properties (383). The pathogenic serotypes are otherwise indistinguishable by morphology or biochemical activity.

Recognition of two species, Leptospira interrogans and L. biflexa, has been proposed for the so-called pathogenic and saprophytic leptospires, respectively (383).

Generally, leptospirosis, typically a disease of animals, has been regarded as an occupational disease that occurs primarily in workers associated with wastewater, rice, sugar cane, farms, and slaughter yards. The disease is transmitted to man by direct contact or via water contaminated by urine from infected wild and domestic animals. Leptospirosis is endemic in some parts of the world. However, 11 outbreaks in nonendemic areas during 1939 to 1959 were associated with swimming or wading in contaminated water (144, 283), indicating the potential for contacting the disease as a result of recreational activity.

At the same time, leptospirosis has become a more recognizable human health problem in recent years, because of better identification of symptoms and improved methods of diagnosis. In rural populations in France, the percentage of infection by source was as follows: water or mud, 21 percent; animals, 53 percent; and water and animals, 25 percent. In nonrural populations, however, 80 percent of the infections was acquired by contact with contaminated water (144). It was also noted that subclinical leptospirosis in humans was not infrequent and may be a significant public health hazard (144). Information on the infective dose of leptospira to humans is not available from the literature surveyed. Leptospira can apparently penetrate intact skin, assisted by cuts, abrasions, and immersion. Incidences of infection from swimming also suggested penetration via the mucous membranes of the mouth or nasopharynx (144). The nesting site of leptospires in natural hosts is the lumen of nephritic tubules, from which they are shed into the urine (144). Pathogenic leptospires can survive for three or more months in neutral or slightly alkaline waters, but do not persist in brackish or acidic water (144). In 1975, two outbreaks of leptospirosis were attributed to swimming in contaminated surface water. Seven children in Tennessee developed infection with L. interrogans serotype grippotyphosa after swimming in a small local stream. Two persons in Louisiana became infected with leptospires of the serotype icterohaemorrhagiae after bathing in a man-made lake (221)

Tuberculosis--

The possibility that tuberculosis may be transmitted by sewage has frequently been considered in connection with the disposal of wastewaters from hospitals, tuberculosis sanitaria, dairies, and slaughter houses; the possibility has even been considered in connection with domestic sewage in general. Concern has been expressed about the danger of human and animal infection, particularly where these waters are reused (266).

The presence of mycobacteria in wastewater has been extensively studied since around 1900 - the time of the first findings of the bacteria in feces (223). The recovery of Mycobacterium tuberculosis (the bacteria that cause tuberculosis) is difficult, even from favorable sources such as sputum; recovery from sewage is much more difficult because of the presence of other bacteria.

The tubercle bacilli are present in the sputum and feces of tuberculosis patients. The wastes from institutions that treat the patients will almost always contain large numbers (4×10^5 to $10^7/\ell$) of tubercle bacilli (266). Significant numbers (about $3/\ell$ in the effluent of a plant producing about 7,600 gpd of milk) of virulent tubercle bacilli are also found in the wastes from some dairies. A cow suffering with tuberculosis of the udders discharges about 1.5×10^8 tubercle bacilli/day (266). M. balnei, which causes granuloma, may be present in chlorinated water used for swimming pools (295). Much of the data on survival of tubercle bacilli in sewage indicate that, under laboratory conditions. the bacteria can be infective for 6 months in sewage and for up to 24 months in feces (266).

Contaminated water can produce typical tuberculosis in humans in some instances. The first clear-cut cases of human infection were reported in 1947. From 1947 to 1953, nine cases of tuberculosis were described in humans who aspirated polluted water into their lungs after swimming and nearly drowning in contaminated water (266).

There were not much data available in the literature on the infectious dose to humans of tubercle bacilli in wastewater. In an experiment with guinea pigs, it was reported that 80 percent of the animals contracted the disease when they were fed with grass that had been sprayed with more than 4×10^6 tubercle bacilli. Calves that had been similarly fed also succumbed (266).

The majority of studies carried out on mycobacteria has focused on the presence of M. tuberculosis in sanitarium wastes. These studies may not provide a realistic picture of the danger of infection from contaminated water.

The potential health hazards of M. tuberculosis being transmitted through sanitarium wastewaters, to drinking supplies, and back to man in general appears to be quite remote.

Viruses--

In the past, transmission of waterborne viral diseases was rarely recognized, due largely to lack of sensitive virus-detection methods and precise quantification. With improved techniques for concentrating viruses from large water samples, increasing occurrences of viruses in water and wastewaters have been reported. Viral transmission through water may take place in various ways: bathing in contaminated water, eating contaminated seafoods, drinking from untreated or improperly treated water sources, or contacting contaminated waters. Enteric viruses have been investigated with greater emphasis than any other group of viruses, mainly because any virus excreted in the feces and capable of producing infection when ingested is theoretically transmissible by water. The human enteric viruses and the diseases associated with them are listed in Tables 110 and 111.

TABLE 110. THE HUMAN ENTERIC VIRUSES AND THE DISEASES ASSOCIATED WITH THEM (142)

Virus Subgroup	No. of Types	Disease
Polio virus	3	paralytic poliomyelitis, aseptic meningitis
Coxsackie virus		
Group A	26	herpangina, aseptic meningitis, paralysis pleurodynia,
Group B	6	aseptic meningitis, acute infantile myocarditis
ECHO virus	34	aseptic meningitis, rash and fever, diarrheal disease, respiratory illnesses
Infectious hepatitis	1(?)	infectious hepatitis
Reovirus	3	fever, respiratory infections, diarrhea
Adenovirus	32	respiratory and eye infections

TABLE 111. THE HUMAN ENTERIC VIRUSES THAT CAN BE WATERBORNE AND KNOWN DISEASES ASSOCIATED WITH THESE VIRUSES (637)

Group	Subgroup	No. of Types or Subtypes	Disease Entities Associated with These Viruses	Pathologic Changes in Patients	Organs Where Virus Multiplies
Enterovirus	Poliovirus	3	Muscular paralysis	Destruction of motor neurons	Intestinal mucosa spinal cord, brain stem
			Aseptic meningitis	Inflammation of meninges from virus	Meninges
			Febrile episode	Viremia and viral multiplication	Intestinal mucosa and lymph
	ECHO virus	34	Aseptic meningitis	Inflammation of meninges from virus	Meninges
			Muscular paralysis	Destruction of motor neurons	Intestinal mucosa spinal cord, brain stem
			Guillain-Barre's Syndrome	Destruction of motor neurons	Spinal cord
			Exanthem	Dilation and rupture of blood vessels	Skin
			Respiratory diseases	Viral invasion of parenchymiatous of respiratory tracts and secondary inflammatory responses	Respiratory tracts and lungs
			Diarrhea	Destruction of intestinal bacteria	Intestine
			Epidemic myalgia	Not well known	
			Pericarditis & myocarditis	Viral invasion of cells with secondary responses	Pericardial and myocardial tissue
			Hepatitis	Viral invasion of cells with secondary responses	Liver parenchyma

TABLE 111. (continued)

Group	Subgroup	No. of Types or Subtypes	Disease Entities Associated with These Viruses	Pathologic Changes in Patients	Organs Where Virus Multiplies
Enterovirus (cont'd)	Coxsackie virus	>24	Herpangina	Viral invasion of mucosa with secondary inflammatory responses	Mouth
	A		Acute lymphatic pharyngitis	Viral invasion of mucosa with secondary inflammatory responses	Lymph nodes and pharynx
			Aseptic meningitis	Inflammation of meninges from virus	Meninges
			Muscular paralysis	Destruction of motor neurons	Intestinal mucosa spinal cord, brain stem
			Hand-foot-mouth disease	Viral invasion of cells of skin of hands and feet and mucosa of mouth	Skin of hands and feet and much of mouth
			Respiratory disease	Viral invasion of parenchyma of respiratory tracts and secondary inflammatory responses	Respiratory tracts and lungs
			Infantile diarrhea	Viral invasion of cells of mucosa	Intestinal mucosa
			Hepatitis	Viral invasion of liver cells	Parenchyma cells of liver
			Pericarditis & myocarditis	Same as before	Same as before
	B	6	Pleurodynia	Viral invasion of muscle cells	Intercostal muscles
			Aseptic meningitis	Same as before	Same as before
			Muscular paralysis	Same as before	Same as before

TABLE 111. (continued)

Group	Subgroup	No. of Types or Subtypes	Disease Entities Associated with These Viruses	Pathologic Changes in Patients	Organs Where Virus Multiplies
Enterovirus (cont'd)	Coxsackie virus B (cont'd)		Meningoencephalitis	Viral invasional invasion of cells	Meninges and brains
			Pericarditis, endocarditis, mycarditis	Same as before	Same as before
			Respiratory diseases	Same as before	Same as before
			Hepatitis or rash	Same as before	Same as before
			Spontaneous abortion	Viral invasion of vascular cells (?)	Placenta
			Insulin-dependent diabetes	Viral invasion of insulin producing cells	Langerhans cells of pancreases
			Congenital heart anomalies	Viral invasion of muscle cells	Developing heart
Reovirus		6	Not well known	Not well known	
Adenovirus		31	Respiratory diseases	Same as before	Same as before
			Acute conjunctivitis	Viral invasion of cells and secondary inflammatory responses	Conjunctival cells and blood vessels
			Acute appendicitis	Viral invasion of mucosa cells	Appendia and lymph nodes
			Intussusception	Viral invasion of lymph nodes (?)	Intestinal lymph nodes (?)
			Sub acute thyroiditis	Viral invasion of parenchyma cells	Thyroid
			Sarcoma in hamsters	Transformation of cells	Muscle cells
Hepatitis		>2	Infectious hepatitis	Invasion of parenchyma cells	Liver

TABLE 111. (continued)

Group	Subgroup	No. of Types or Subtypes	Disease Entities Associated with These Viruses	Pathologic Changes in Patients	Organs Where Virus Multiplies
Hepatitis (cont'd)			Serus hepatitis	Invasion of parenchyma cells	Liver
			Down's Syndrome	Invasion of cells	Frontal lobe of brain, muscle, bones

Infectious Hepatitis--

Apart from theoretical considerations, there are very few viruses for which epidemiological evidence suggests transmission by water. Infectious hepatitis (hepatitis A) is the only disease caused by an agent having the characteristics of a virus for which evidence of waterborne transmission has been accepted by all workers in the field(468). Therefore, it is regarded as the viral disease of greatest importance in wastewater.

In 1973 alone, a total of 59,200 cases of viral hepatitis A, B, and a type unspecified were reported (221). In 1974, a total of 59,340 cases of viral hepatitis - hepatitis A, B, and type unspecified - were reported to CDC. This represents a rate of 28.1 cases/100,000 population, approximately the same rate as for 1973. Since 1971, 1974 is the first year to have shown rate increases for two quarters; the increase in cases began in the fourth quarter of 1973. The seasonal variation noted in the 1950s and early 1960s was not seen in 1974. The 48,709 cases of acute hepatitis A and hepatitis, type unspecified, constituted 82.1 percent of the total cases of viral hepatitis reported in 1974 (291). Waterborne outbreaks of hepatitis A continue to occur in the United States. From 1971 to 1973, these documented outbreaks were associated with contaminated drinking water from either municipal, semipublic, or individual water systems (144, 448). Use of contaminated spring or groundwater without proper treatment or disinfection, and back-siphonage of contaminated water into the distribution system were reported to be the causes of the outbreaks. The majority of documented hepatitis A outbreaks in municipal water systems in the United States between 1946 and 1971 occurred as a result of distribution system contamination, primarily through cross connections and back-siphonage.

Two outbreaks of shellfish-associated hepatitis involving 285 cases were reported in 1973 (221). Both outbreaks - one in Georgia and the other in Texas - were associated with the eating of raw oysters. Epidemiologic evidence suggested that two particular bays contaminated by flooding were the source of the contaminated oysters.

A recent hepatitis outbreak of 14 cases was reported to be associated with swimming in a grossly contaminated lake in North Carolina, and with ingesting water from that lake. This is the first time that such a definite case has been made for the potential of contracting this disease while swimming in sewage-polluted water. The probable fecal-oral transmission of infectious hepatitis made the waterborne route possible. This mode of transmission was vividly illustrated by several large epidemics that took place in 1955 to 1956, especially in India where 28,745 cases occurred (468).

As far as the magnitude of waterborne infectious hepatitis is concerned, the water route still only accounts for up to 1 percent of reported cases at any time for which information is available (468).

Despite the increased interest and concern in infectious hepatitis, its infectious agent has not yet been isolated and cultured. One recent report (212) using microscopic techniques was able to show the presence of viruslike particles, immunologically distinct from hepatitis B, in infected stools.

Poliomyelitis--

The infectivity of feces from persons with poliomyelitis and the characteristic fecal excretion of the diseased persons have been documented for years. The polio virus has been sought and detected in sewage. Accordingly, the water route of transmission has been implicated in several outbreaks of poliomyelitis (see Table 112). Many cases of epidemics of poliomyelitis were attributed to waterborne transmission through contaminated or untreated water, but the evidence is not sufficient. It appears that water transmission of the polio virus may be a rare occurrence in the United States, but common in parts of the world lacking adequate sanitary facilities. Six of the outbreaks attributed to drinking water occurred in Sweden during the 1930s and 1940s (468) and led to the early recognition of the importance of the fecal-oral route in poliomyelitis.

Viral Gastroenteritis--

When a recognized pathogen cannot be isolated in cases of gastroenteritis and diarrhea, the term viral is often used to describe the symptoms. It is quite possible that forms of gastroenteritis and diarrhea transmissible from person to person are due to viruses.

Gastroenteritis and diarrheal disease are believed to have accounted for approximately 60 percent of all epidemics of waterborne diseases throughout history. The number of these cases that was due to viral agents is not known; if, however, only a small portion of the cases was due to viral agents, this number would still be quite substantial.

A viruslike particle, similar in appearance but immunologically distinct from the hepatitis A, has been reported to be associated with an acute infectious nonbacterial gastroenteritis (341). Shellfish-associated gastroenteritis has also been reported (177).

TABLE 112. PUBLISHED REPORTS OF POLIOMYELITIS ATTRIBUTED TO CONTAMINATED DRINKING WATER(468)

No.	Yr of occurrence	References	Country	Place or type of population	No. cases attributed to supply
1	1913(?)-39	Spaak, 1941	Sweden	Rural district	10[b]
2	1944	Kling, 1947	Sweden	Town	63
3	1944	Kling, 1947	Sweden	Town	53
4	1948	Faahraeus et al., 1950	Sweden	Stockholm suburb	9
5	1948	Faahraeus et al., 1950	Sweden	Town	63[c]
6	1949	Huss et al., 1952	Sweden	Malmoe	138
7	1952	Bancroft et al., 1957	U.S.A.	"Huskerville"	45
8	1953	Little, 1954	Canada	Edmonton	75[b]

a Unadjusted rate among persons presumably consuming contaminated supply.

b Estimated from author's data.

c Includes cases attributed to other modes of transmission.

TABLE 112 . (continued)

No.	Attack rate per 100[a]	Character of Episode	Duration of waterborne phase	Type of supply held responsible
1	--	Sporadic cases	Yrs	Private well, pond
2	0.5	Epidemic	5 mo	Municipal system Filtered surface water
3	0.2	Epidemic	3 mo	Municipal system Untreated deep well water
4	--	Sporadic cases	3 mo	Municipal system Proximate contamination
5	0.2	Epidemic	7 mo	Municipal system Untreated deep well water
6	0.1	Epidemic	6 mo	Municipal system Filtered surface water
7	6.7	Epidemic	5 weeks	Municipal system Proximate contamination
8	<0.1[b]	Epidemic	2 weeks	Municipal system Chlorinated surface water

a Unadjusted rate among persons presumably consuming contaminated supply.

b Estimated from author's data.

c Includes cases attributed to other modes of transmission.

Infective Dose of Viruses--

Virologists feel that one plaque-forming unit (pfu) - one viral particle that grows in the laboratory media - constitutes an infectious viral dose. However, there is a difference between infection and disease (637); the diseased person manifests a variety of symptoms and readily recognizes that he is sick; an infected person has the material in his system, but does not necessarily show symptoms of the disease. It has been estimated that of every 100 to 1,000 people who are infected, only one will manifest the clinical symptoms of disease. However, it is not quite as simple as these statistics suggest, for an infected individual can serve as a carrier or source within the community and transmit this disease to other people (637).

Table 113 shows the minimal infective doses of attenuated polio virus for human hosts by oral routes. The infected rate with 20 or more pfu was 100 percent; with 2 pfu, 67 percent of the individuals was infected.

TABLE 113. MINIMAL INFECTIVE DOSES OF ATTENUATED POLIO VIRUSES FOR HUMAN HOSTS BY ORAL ROUTES (637).

Subject	Virus	Dose (pfu)	Carrier* Rate	Infected Percent
Adults	Polio virus Type I	200	4/4	100
	(SM Strain)	20	4/4	100
		2	2/3	67
		0.2	0/2	0
Premature Infants	Polio virus Type III	10	2/3	67
	(Fox Strain)	2.5	3/9	33
		1	3/10	30

* Number of persons developed into carrier for the virus/number of persons who had taken the virus orally.

It has been mentioned that infection with small amounts of virus in water would probably immunize individuals rather than produce disease. This may be substantiated by the fact that sewage workers continually exposed to small amounts of infected material had the lowest rate of absenteeism among all the occupation groups studied (52).

An adequate biological indicator for viruses in various waters is not currently available; however, efforts are being made to find better indicators. For example, ratios of coliphages to human enteric viruses and the coliform-virus ratio have been investigated (235, 364) Also, a high degree of coliforms and coliphage occurring in water samples (346) as well as a yeast and two acid-fast bacilli recovered from wastewater, resist chlorination at a level sufficient to inactivate viruses. This suggests that they may be useful indicators of wastewater chlorination efficiency (201).

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